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PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference 55791W0007	FOR FURTHER ACTION see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. PCT/US 01/ 24867	International filing date (day/month/year) 08/08/2001	(Earliest) Priority Date (day/month/year) 23/08/2000
Applicant 3M INNOVATIVE PROPERTIES COMPANY		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

1. Basis of the report

- a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ **Certain claims were found unsearchable** (See Box I).

3. ☐ **Unity of invention is lacking** (see Box II).

4. With regard to the **title**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

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5. With regard to the **abstract**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No.

☒ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

1
☐ None of the figures.

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INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 01/24867

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 B29C47/06 B32B1/08 B32B27/08 F16L9/12 F16L11/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 B29C B32B F16L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 641 445 A (DUKES GLENN V ET AL) 24 June 1997 (1997-06-24) column 1, line 15 -column 2, line 23 column 2, line 52 -column 3, line 55 column 5, line 63 -column 6, line 1 column 6, line 46 - line 56; claims 1,2,10,20-22	1,2, 8-10, 12-19, 24,28
X A	column 6, line 56 -column 7, line 56 claims 19,23-25 ---	21,23 29
A	WO 96 00657 A (DUKES GLENN V ;BROWDER WILLIAM TROY (US); CADILLAC RUBBER & PLASTI) 11 January 1996 (1996-01-11) claims 1,7,11,14,18,19 ---	1,2, 8-10, 12-19, 21,23, 24,28,29
	--- -/--	



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

* Special categories of cited documents :

- *A* document defining the general state of the art which is not considered to be of particular relevance
- *E* earlier document but published on or after the international filing date
- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

- *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- *&* document member of the same patent family

Date of the actual completion of the international search

5 October 2001

Date of mailing of the international search report

12/10/2001

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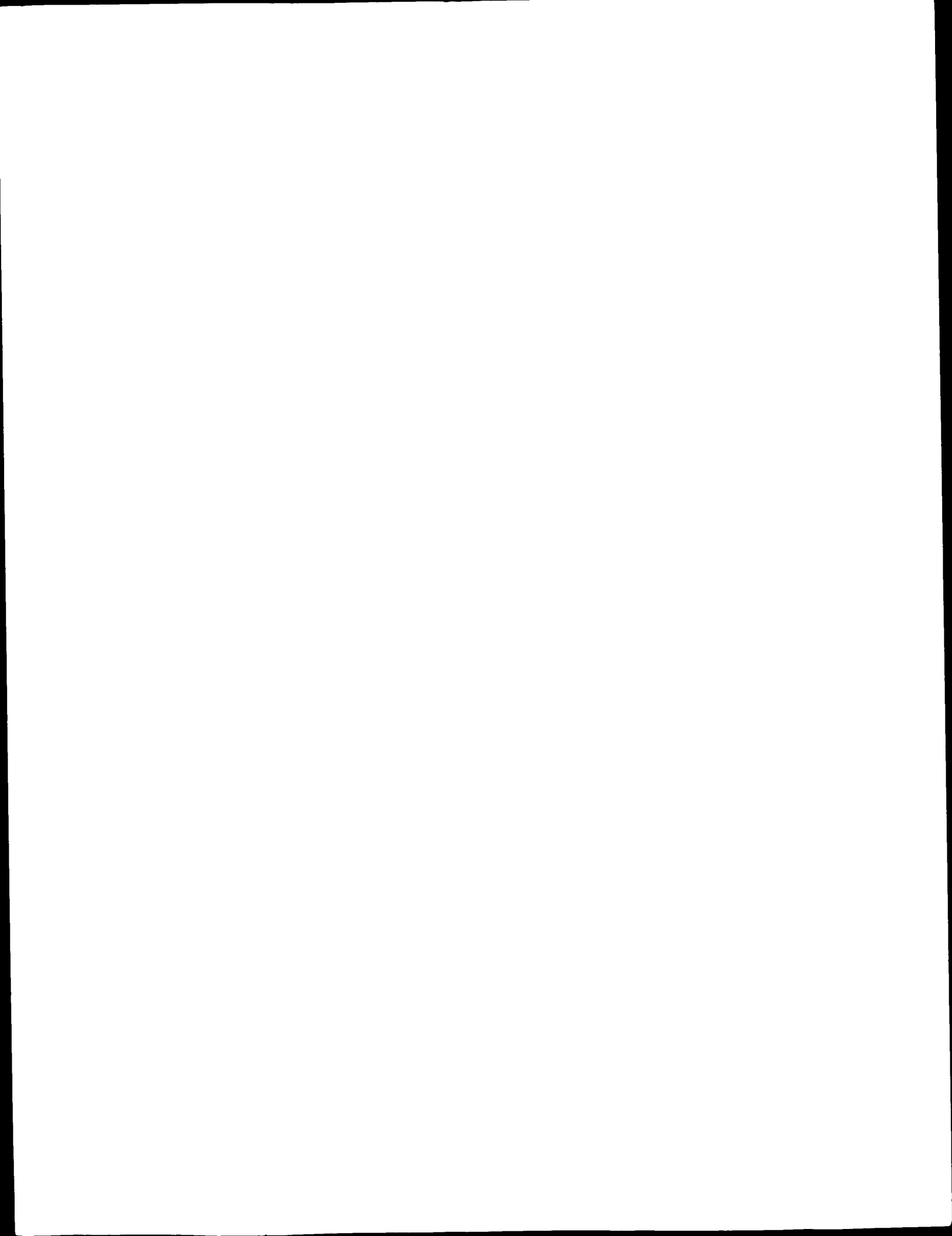
INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 01/24867

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 551 094 A (PILOT IND INC) 14 July 1993 (1993-07-14) figures 3-21 ----	1,28
A	WO 99 32557 A (DYNEON LLC) 1 July 1999 (1999-07-01) page 1, line 10 - line 14 page 3, line 23 -page 8, line 29 page 9, line 26 -page 11, line 30 A page 13, line 30 -page 14, line 10 examples 1-8; tables 1,2 ----	10-20, 24,25
A	US 4 895 744 A (BRIGGS MILTON ET AL) 23 January 1990 (1990-01-23) column 1, line 64 -column 3, line 17; figure 4 column 11, line 22 - line 40 ----	21-23,29
A	PATENT ABSTRACTS OF JAPAN vol. 013, no. 036 (M-790), 26 January 1989 (1989-01-26) & JP 63 246224 A (SUMITOMO ELECTRIC IND LTD), 13 October 1988 (1988-10-13) abstract -----	1,3,4



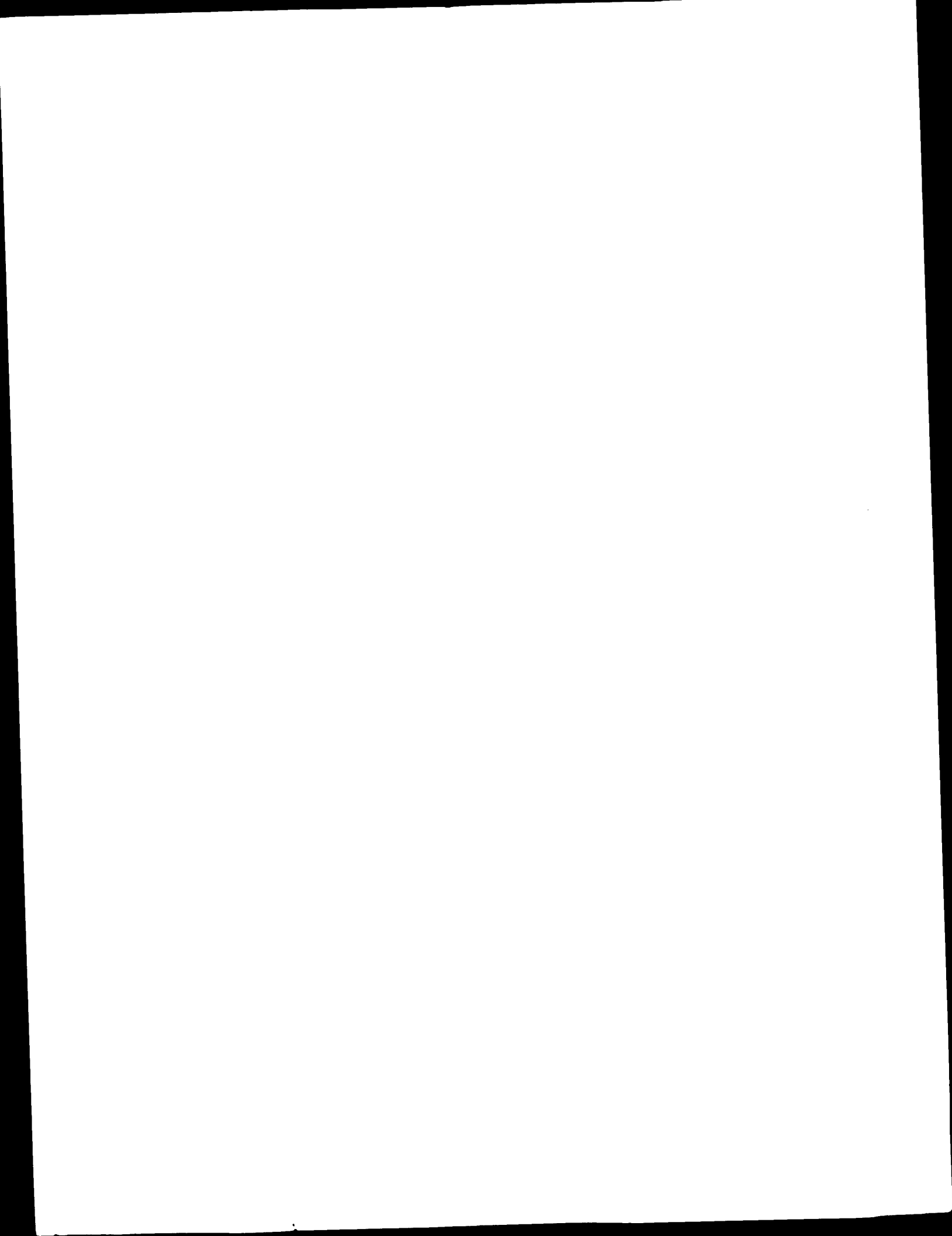
INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 01/24867

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 5641445	A	24-06-1997	WO 9846412 A1 AU 2733397 A BR 9714610 A DE 980306 T1 EP 0980306 A1	22-10-1998 11-11-1998 23-05-2000 05-10-2000 23-02-2000
WO 9600657	A	11-01-1996	WO 9600657 A1 DE 69426120 D1 DE 69426120 T2 EP 0716632 A1 ES 2153426 T3 US 5941286 A	11-01-1996 16-11-2000 10-05-2001 19-06-1996 01-03-2001 24-08-1999
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JP 63246224	A	13-10-1988	NONE	



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NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Commissioner
 US Department of Commerce
 United States Patent and Trademark
 Office, PCT
 2011 South Clark Place Room
 CP2/5C24
 Arlington, VA 22202
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Date of mailing (day/month/year)

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Priority date (day/month/year)

23 August 2000 (23.08.00)

Applicant

FUKUSHI, Tatsuo et al

1. The designated Office is hereby notified of its election made:



in the demand filed with the International Preliminary Examining Authority on:

04 December 2001 (04.12.01)



in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was

was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

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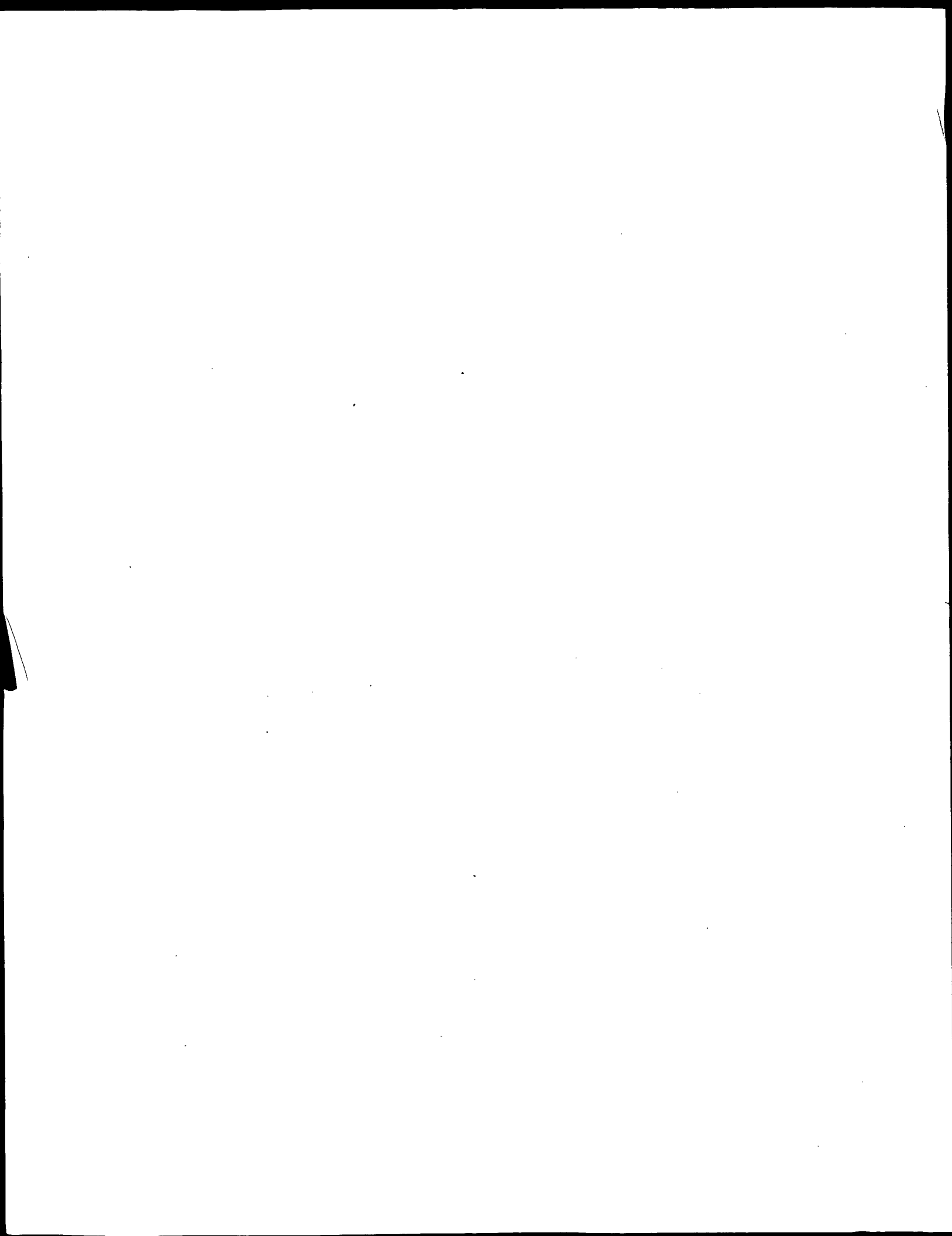
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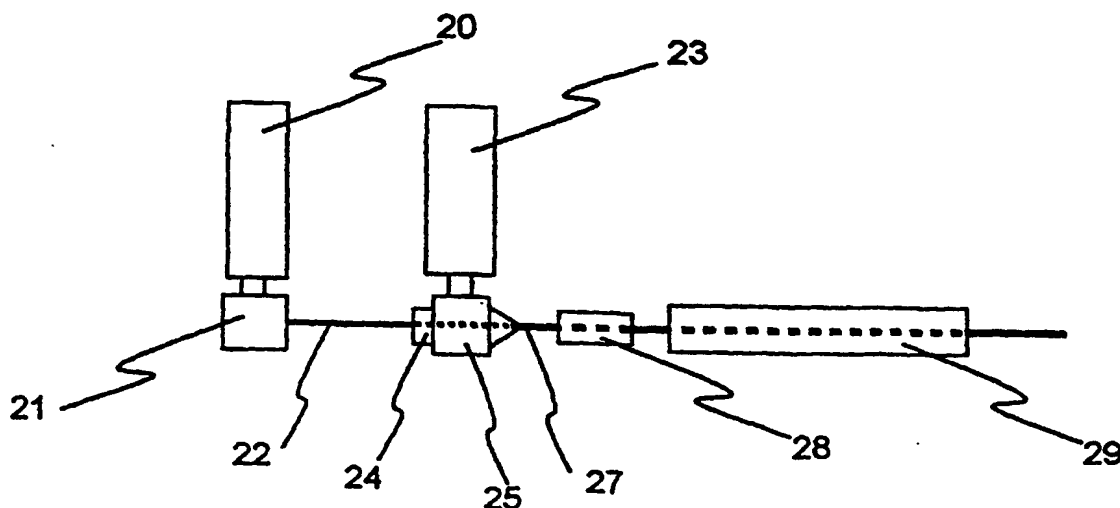
(74) Agents: LILLY, James, V. et al.; Office of Intellectual
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55133-3427 (US).

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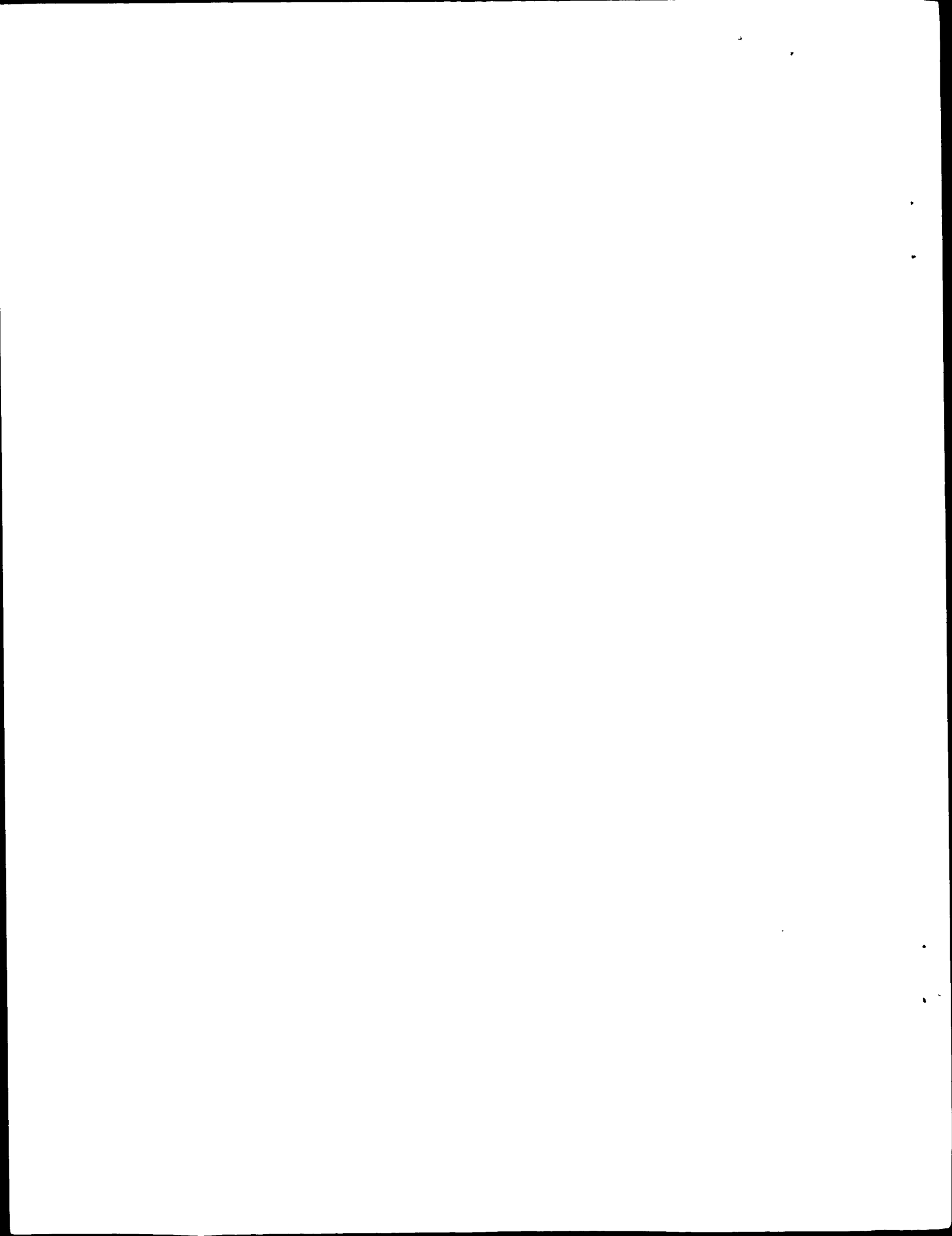
[Continued on next page]

(54) Title: PROCESS FOR PREPARING A MULTI-LAYER ARTICLE HAVING A FLUOROPLASTIC LAYER AND AN ELAS-
TOMER LAYER



(57) Abstract: A method for enhancing the bond strength between a VDF-containing fluoroplastic layer and an elastomer layer of a multi-layer article. A VDF-containing fluoroplastic composition is applied to the surface of a precursor article that includes a curable elastomer layer to form a fluoroplastic layer. Prior to application of the fluoroplastic composition, the curable elastomer layer is thermally insulated to prevent it from undergoing substantial heating. Following application, the fluoroplastic layer is heated and the curable elastomer layer is cured (e.g., thermally cured). Preferably, the elastomer cure occurs separately from and subsequent to heating the fluoroplastic layer. The combination of thermally insulating the curable elastomer layer prior to application of the fluoroplastic composition and heating the fluoroplastic layer following application of the fluoroplastic composition results in formation of a strong bond between the fluoroplastic and elastomer layers upon cure.

WO 02/16112 A1



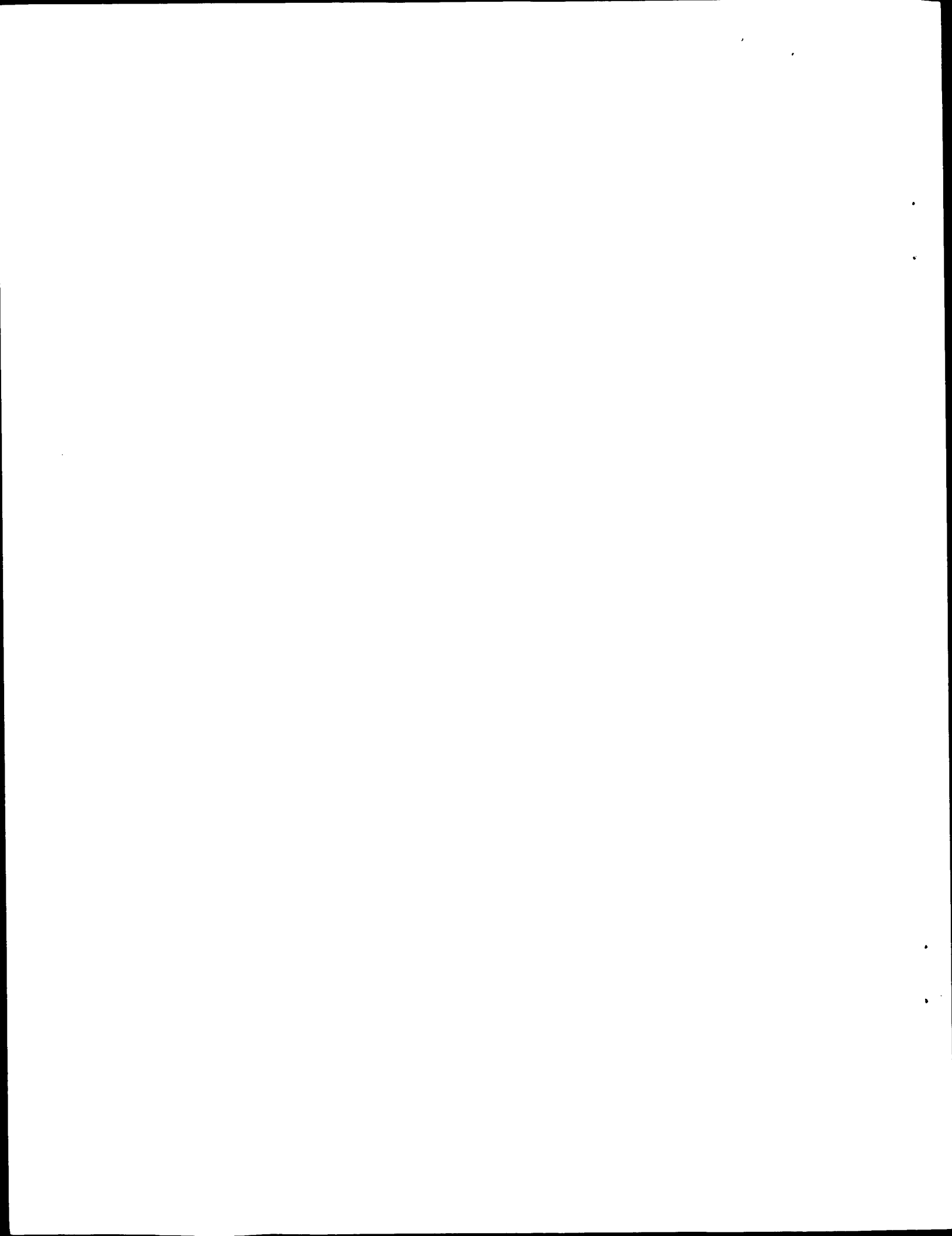


patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

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PROCESS FOR PREPARING A MULT-LAYER ARTICLE HAVING A FLUOROPLASTIC LAYER AND AN ELASTOMER LAYER

TECHNICAL FIELD

This invention relates to preparing multi-layer articles having a fluoroplastic layer and
5 an elastomer layer.

BACKGROUND

Fluorine-containing polymers (also known as "fluoropolymers") are a commercially useful class of materials. Fluoropolymers include, for example, crosslinked fluoroelastomers and semi-crystalline or glassy fluoroplastics. Fluoroplastics are generally of high thermal
10 stability and are particularly useful at high temperatures. They may also exhibit extreme toughness and flexibility at very low temperatures. Many of these fluoroplastics are almost totally insoluble in a wide variety of solvents and are generally chemically resistant. Some have extremely low dielectric loss and high dielectric strength, and may have unique non-adhesive and low friction properties. See, e.g., F.W. Billmeyer, *Textbook of Polymer Science*,
15 3d ed., pp. 398-403, John Wiley & Sons, New York (1984).

Fluoroelastomers, particularly the copolymers of vinylidene fluoride with other ethylenically unsaturated halogenated monomers such as hexafluoropropylene, have particular utility in high temperature applications such as seals, gaskets, and linings. See, e.g., R.A. Brullo, "Fluoroelastomer Rubber for Automotive Applications," *Automotive Elastomer &*
20 *Design*, June 1985; "Fluoroelastomer Seal Up Automotive Future," *Materials Engineering*, October 1988; and W.M. Grootaert et al., "Fluorocarbon Elastomers," Kirk-Othmer, *Encyclopedia of Chemical Technology*, vol. 8, pp. 990-1005 (4th ed., John Wiley & Sons, 1993).

Multi-layer constructions containing a fluoropolymer enjoy wide industrial
25 application. Such constructions find utility, for example, in fuel line hoses and related containers and hoses or gaskets in the chemical processing field. Increased concerns with evaporative fuel standards give rise to a need for fuel system components that have increased barrier properties to minimize the permeation of fuel or fuel vapors through automotive components such as fuel filler lines, fuel supply lines, fuel tanks, and other components of the

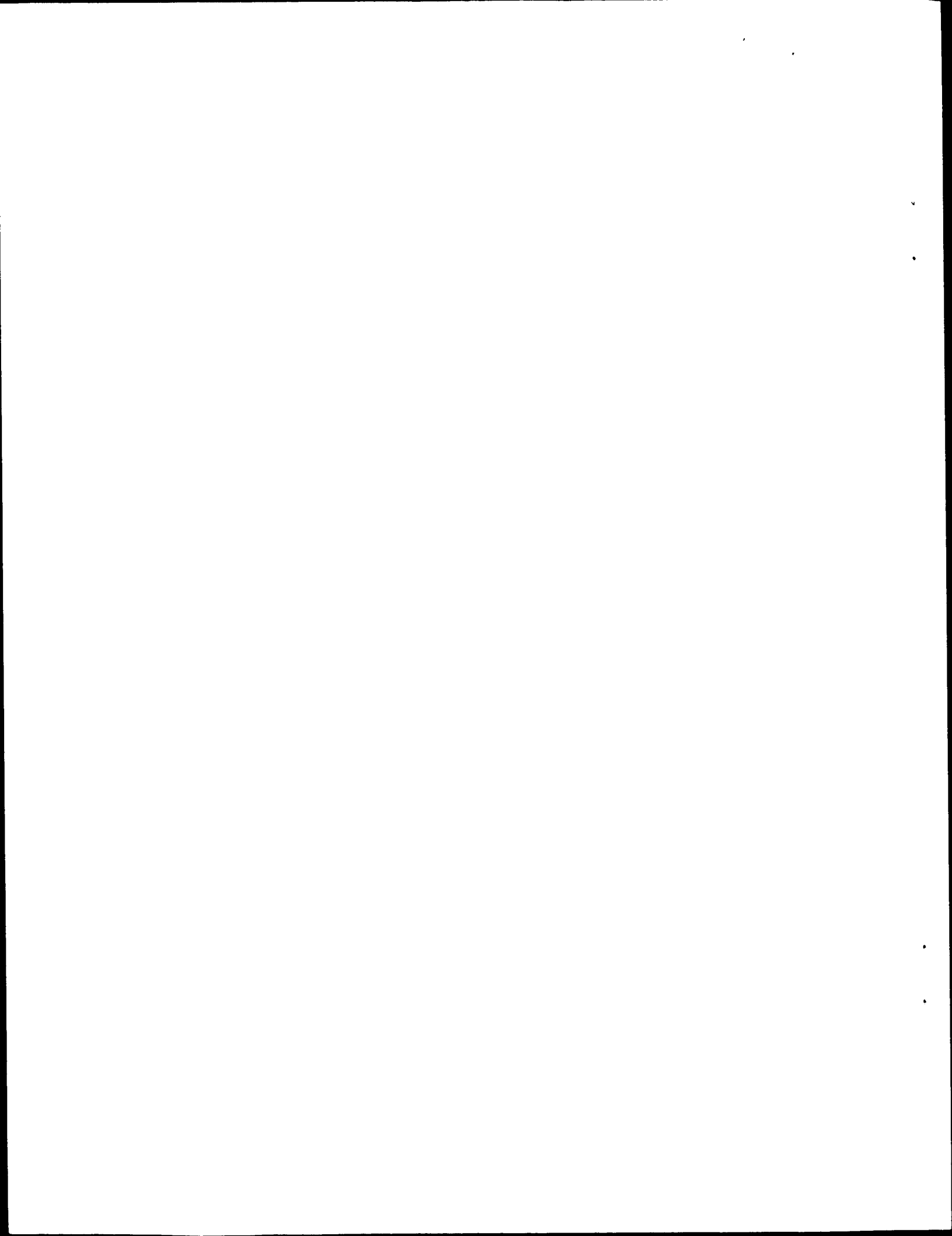
engine's fuel or vapor recovery systems. Various types of tubing have been proposed to address these concerns.

Adhesion between the layers of a multi-layered article may need to meet various performance standards depending on the use of the finished article. However, it is often difficult to establish high bond strengths when one of the layers is a fluoropolymer. Various methods have been proposed to address this problem. One approach is to use an adhesive layer or tie layer between the fluoropolymer layer and the second polymer layer. Surface treatments for the fluoropolymer layer, including solvent etching and corona discharge, have also been employed to enhance adhesion. In the case of fluoropolymers containing interpolymers derived from vinylidene fluoride, exposure of the fluoropolymer to a dehydrofluorinating agent such as a base has been used, as well as polyamine reagents applied to the fluoropolymer surface or incorporated within the fluoropolymer itself.

SUMMARY

The invention relates to a method for enhancing the bond strength between a fluoroplastic layer and an elastomer layer of a multi-layer article. The elastomer may be a fluoroelastomer or a non-fluorinated elastomer. According to the method, a fluoroplastic composition that includes interpolymers derived from vinylidene fluoride (VDF) is applied to the surface of a precursor article that includes a curable elastomer layer, preferably by extrusion coating the composition in molten form through a crosshead die, to form a fluoroplastic layer. Preferably, the composition is applied directly to the surface of the elastomer layer. Prior to application of the fluoroplastic composition, the curable elastomer layer is thermally insulated to prevent it from undergoing substantial heating. In one embodiment, where molten fluoroplastic composition is extrusion coated through a crosshead die, thermal insulation is achieved by equipping the die with a sleeve located at least partially within the upstream end of the die that receives and thermally insulates the curable elastomer layer prior to application of the fluoroplastic composition.

Following application, the fluoroplastic layer is heated and the curable elastomer layer is cured (preferably thermally cured). Preferably, the elastomer cure occurs separately from and subsequent to heating of the fluoroplastic layer. The combination of thermally insulating the curable elastomer layer prior to application of the fluoroplastic composition and heating



the fluoroplastic layer following application of the fluoroplastic composition results in formation of a strong bond between the fluoroplastic and elastomer layers upon cure, even in the absence of adhesion aids such as surface treatments, separate adhesive layers, and the like. For example, bond strengths of at least 15 N/cm, can be achieved.

5 Multi-layer articles prepared according to this method can be provided in a wide variety of shapes, including sheets, films, containers, hoses, tubes, and the like. The articles are especially useful wherever chemical resistance and/or barrier properties are necessary. Examples of specific uses for the articles include their use in rigid and flexible retroreflective sheets, adhesive articles such as adhesive tapes, paint replacement films, drag reduction films,
10 fuel line and filler neck hoses, exhaust handling hoses, fuel tanks, and the like. The articles are also useful in chemical handling and processing applications, and as wire and cable coatings or jackets.

The details of one or more embodiments of the invention are set forth in the accompanying drawings and the description below. Other features, objects, and advantages of
15 the invention will be apparent from the description and drawings, and from the claims.

DESCRIPTION OF DRAWINGS

FIG. 1 is a schematic drawing of a process for making a multi-layered article according to the invention.

Like reference symbols in the various drawings indicate like elements.

20

DETAILED DESCRIPTION

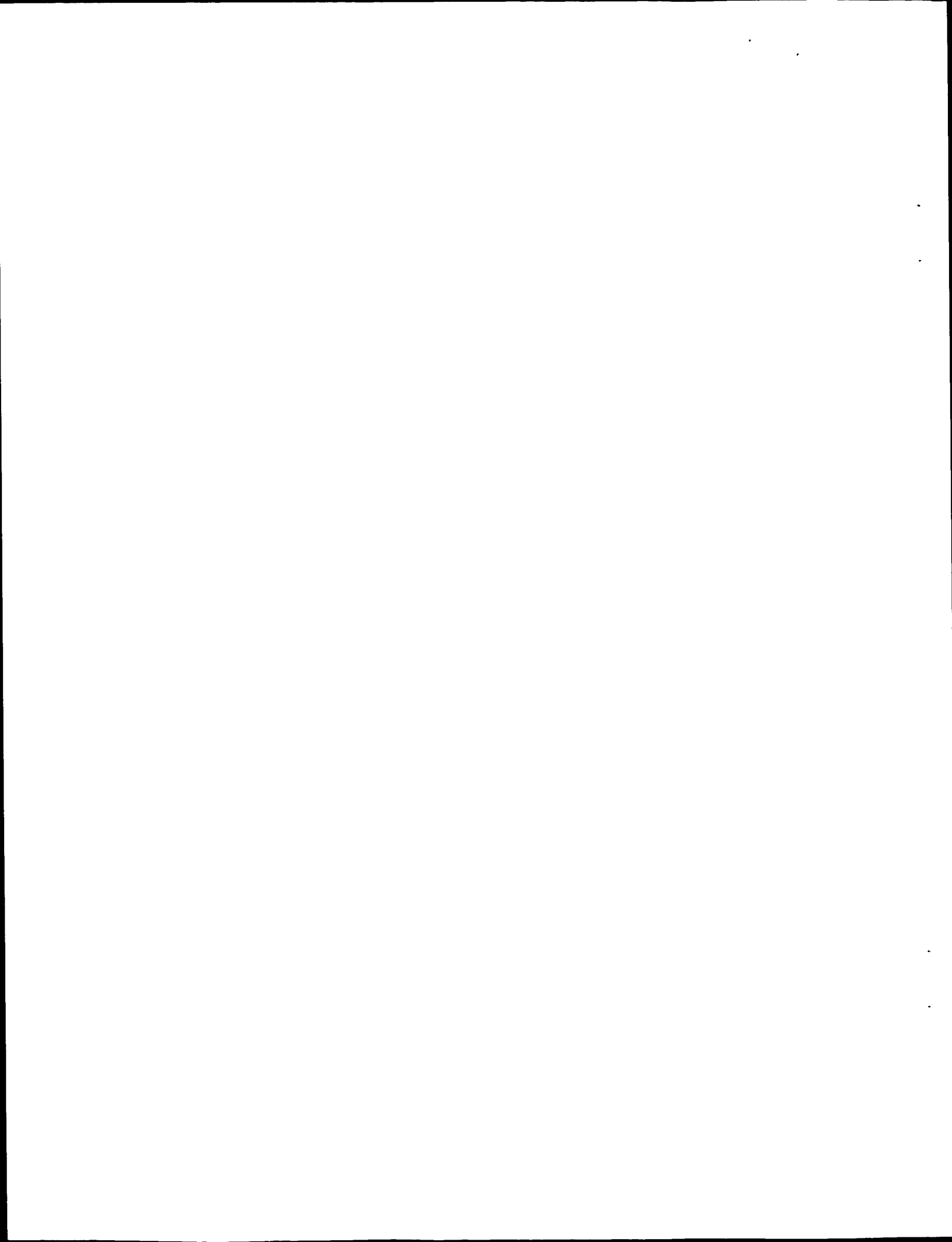
Referring to Fig. 1, there is shown one embodiment of a process for preparing a multi-layer article featuring a fluoroplastic layer bonded to an elastomer layer. An extruder 20 extrudes a curable elastomer composition through a die 21 to form a length of tubing 22 having a curable elastomer layer. A second extruder 23 located downstream of extruder 20
25 and equipped with a crosshead die 25 coats a layer of molten fluoroplastic onto the surface of the curable elastomer layer. A plastic (or other thermally insulating material) sleeve 24, e.g., a tetrafluoroethylene sleeve, inserted partially within the upstream opening of die 25 receives tubing 22 and thermally insulates it prior to extrusion coating, thereby preventing substantial heating of the curable elastomer layer prior to application of the fluoroplastic. The absence of

substantial heating prior to application of the fluoroplastic contributes to the development, upon cure, of a strong bond between the fluoroplastic and elastomer layers. It may also be desirable to cool the curable elastomer prior to application of the fluoroplastic. This may be accomplished, for example, by treating the curable elastomer layer with a solvent that could
5 then be removed by evaporation.

Following extrusion coating, the resulting multi-layer article 27, featuring a fluoroplastic layer deposited on a curable elastomer layer, enters a tubular heater 28 that heats the fluoroplastic layer. An example of a useful tubular heater is a radiant heater. During the heating process, heat is transferred from heater 28 to the fluoroplastic layer, and then
10 transferred inwardly from the fluoroplastic layer to the curable elastomer layer. It is believed that this heating step contributes to the development, upon cure, of a strong bond between the fluoroplastic and elastomer layers. Following the heating operation, the multi-layer article may be cooled, e.g., by immersion in a cooling bath 29.

The elastomer layer may be cured either in heater 28, or, more preferably, in a
15 separate step under pressure and higher temperature either before or after immersion in cooling bath 29. For example, it may be desirable to cool the article in bath 29, cut it into appropriately sized pieces, and then heat the individual pieces under pressure, e.g., in an autoclave, to cure the curable elastomer layer.

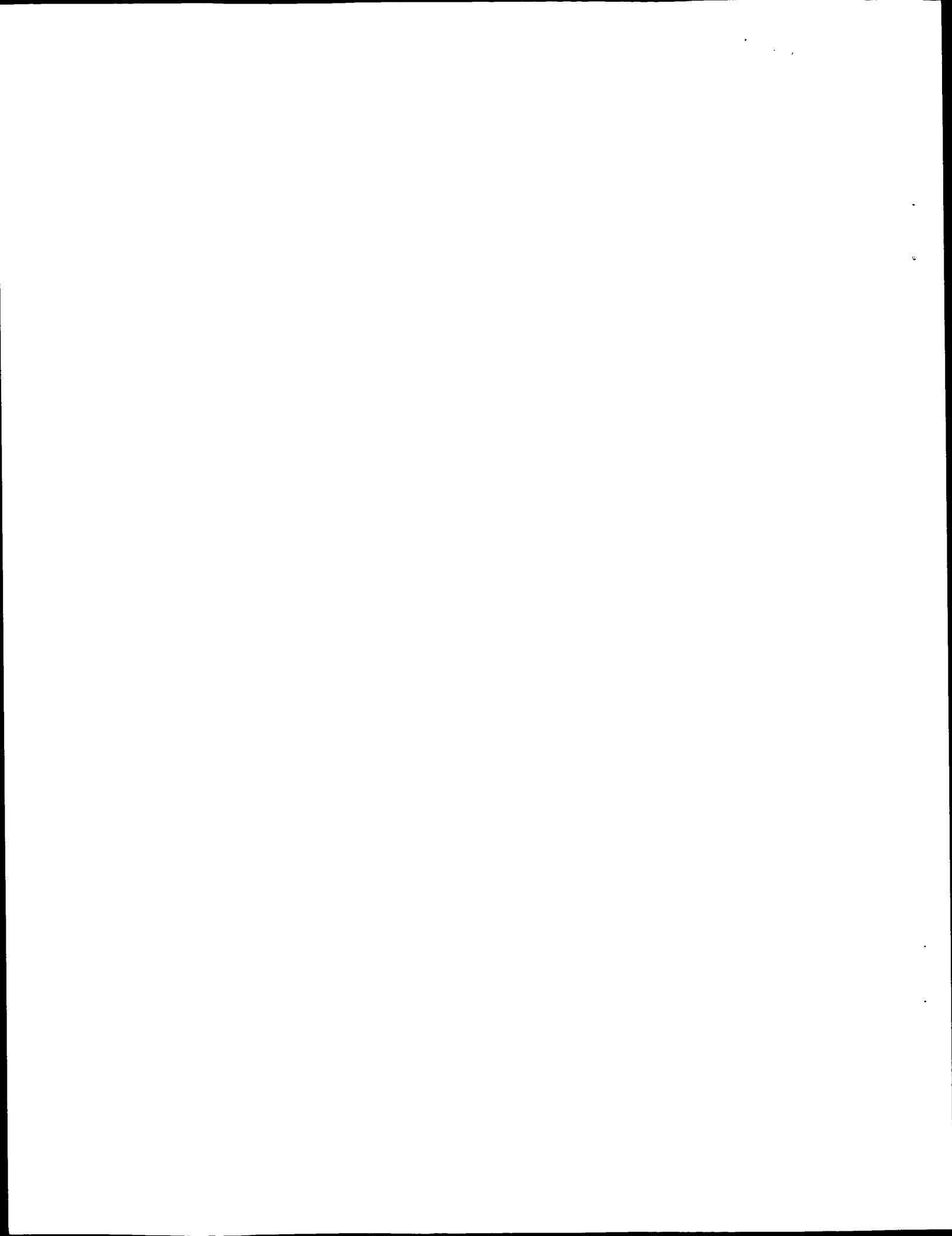
The fluoroplastic preferably is a material that is capable of being extrusion coated.
20 Such fluoroplastics typically have melting temperatures ranging from about 100 to about 330°C, more preferably from about 150 to about 270°C. The fluoroplastic includes interpolymerized units derived from VDF and may further include interpolymerized units derived from other fluorine-containing monomers, non-fluorine-containing monomers, or a combination thereof. Examples of suitable fluorine-containing monomers include
25 tetrafluoroethylene (TFE), hexafluoropropylene (HFP), chlorotrifluoroethylene (CTFE), 3-chloropentafluoropropene, perfluorinated vinyl ethers (e.g., perfluoroalkoxy vinyl ethers such as $\text{CF}_3\text{OCF}_2\text{CF}_2\text{CF}_2\text{OCF}=\text{CF}_2$ and perfluoroalkyl vinyl ethers such as $\text{CF}_3\text{OCH}=\text{CF}_2$ and $\text{CF}_3\text{CF}_2\text{CF}_2\text{OCF}=\text{CF}_2$), and fluorine-containing di-olefins such as perfluorodiallylether and perfluoro-1,3-butadiene. Examples of suitable non-fluorine-containing monomers include
30 olefin monomers such as ethylene, propylene, and the like.



The VDF-containing fluoroplastics may be prepared using emulsion polymerization techniques as described, e.g., in Sulzbach et al., U.S. 4,338,237, hereby incorporated by reference. Useful commercially available VDF-containing fluoroplastics include, for example, THV 200, THV 400, THV 500G, THV 610X fluoropolymers (available from
5 Dyneon LLC, St. Paul, MN), KYNAR 740 fluoropolymer (available from Atochem North America, Philadelphia, PA), HYLAR 700 (available from Ausimont USA, Inc., Morristown, NJ), and FLUOREL FC-2178 (available from Dyneon LLC).

A particularly useful fluoroplastic includes interpolymers derived from at least TFE and VDF in which the amount of VDF is at least 0.1% by weight, but less than 20%
10 by weight. Preferably, the amount of VDF ranges from 3-15% by weight, more preferably from 10-15% by weight.

The curable elastomer may be a fluoroelastomer or a non-fluorinated elastomer. Examples of suitable fluoroelastomers include VDF-HFP copolymers, VDF-HFP-TFE terpolymers, TFE-propylene copolymers, and the like. Examples of suitable non-fluorinated
15 elastomers include acrylonitrile butadiene (NBR), butadiene rubber, chlorinated and chlorosulfonated polyethylene, chloroprene, ethylene-propylene monomer (EPM) rubber, ethylene-propylene-diene monomer (EPDM) rubber, epichlorohydrin (ECO) rubber, polyisobutylene, polyisoprene, polysulfide, polyurethane, silicone rubber, blends of polyvinyl chloride and NBR, styrene butadiene (SBR) rubber, ethylene-acrylate copolymer rubber, and
20 ethylene-vinyl acetate rubber. Commercially available elastomers include NipolTM 1052 NBR (Zeon Chemical, Louisville, KY), HydrinTM C2000 epichlorohydrin-ethylene oxide rubber (Zeon Chemical, Louisville, KY), HypalonTM 48 chlorosulfonated polyethylene rubber (E.I. DuPont de Nemours & Co., Wilmington, DE), NordelTM EPDM (R.T. Vanderbilt Co., Inc., Norwalk, CT), VamacTM ethylene-acrylate elastomer (E.I. DuPont de Nemours & Co.
25 Wilmington, DE), KrynacTM NBR (Bayer Corp., Pittsburgh, PA), PerbunanTM NBR/PVC blend (Bayer Corp., Pittsburgh, PA), TherbanTM hydrogenated NBR (Bayer Corp., Pittsburgh, PA), ZetpolTM hydrogenated NBR (Zeon Chemical, Louisville, KY), SantopreneTM thermoplastic elastomer (Advanced Elastomer Systems, Akron, OH), and KeltanTM EPDM (DSM Elastomers Americas, Addis, LA).



A curing agent is preferably blended with the curable elastomer to facilitate cure. Examples of useful curing agents include imidazolines, diamines, internal salts of diamines, thioureas, and polyphenol curing agents as discussed in U.S. 4,287,322 (Worm), incorporated herein by reference. Such agents are particularly useful for epichlorohydrin compositions.

5 Other examples, particularly useful in the curing of nitrile rubber-containing compositions, include peroxide compounds and sulfur-containing compounds.

In the case of curable fluoroelastomers, examples of useful curing agents include polyols in combination with organo-onium salts (e.g., organo-ammonium, organo-phosphonium, and organo-sulfonium salts). Specific examples are described, e.g., in Fukushi,
10 U.S. 5,658,671, "Fluoroelastomer Coating Composition," hereby incorporated by reference. Diamines and peroxides are also useful.

The multi-layer article may contain additional polymer layers as well. Examples of suitable polymer layers include non-fluorinated polymers such as polyamides, polyimides, polyurethanes, polyolefins, polystyrenes, polyesters, polycarbonates, polyketones, polyureas,
15 polyacrylates, and polymethylmethacrylates. Adhesion between a fluorothermoplastic layer, a fluoroelastomer layer and an elastomer layer can be improved by step curing the three extruded layers in which the elastomer layer is an outside layer, the fluoroplastic layer is a middle layer and the fluoroelastomer layer is an inside layer.

A particularly useful construction for fuel applications features a relatively thin layer
20 of the fluoroplastic that acts as a barrier layer bonded on one face to a relatively thick layer of non-fluorinated polymer that acts as a coverstock, and on the opposite face to a relatively thin elastomer layer (e.g., a fluoroelastomer or a non-fluorinated elastomer) that performs a sealing function. The coverstock provides the article with structural integrity. To further enhance structural integrity, reinforcing aids such as fibers, mesh, and/or a wire screen may be
25 incorporated in the multi-layer article, e.g., as separate layers or as part of an existing layer.

Any or all of the individual layers of the multi-layer article may further include one or more additives. Examples of useful additives include pigments, plasticizers, tackifiers, fillers, electrically conductive materials (e.g., of the type described in U.S. 5,552,199), electrically insulating materials, stabilizers, antioxidants, lubricants, processing aids, impact modifiers,
30 viscosity modifiers, and combinations thereof. For example, in the case of the multi-layer

article for fuel applications described above, it is often useful for the innermost layer of the construction to be electrically conductive.

In some cases, it may be desirable to further enhance bond strength between individual layers of the multi-layer article. For example, the article may be subjected to additional heat,
5 pressure, or both, following cure.

Another way of increasing the bond strength between the layers is to treat the surface of one or more of the layers prior to forming the multi-layered articles. Such surface treatments may consist of a solution treatment using a solvent. If the solvent contains a base, e.g., 1,8-diaza[5.4.0]bicyclo undec-7-ene (DBU), treatment of the fluoropolymer will result in
10 some degree of dehydrofluorination. Such dehydrofluorination may be beneficial to promote adhesion to subsequently applied materials. This is particularly true when the subsequently applied material contains any agent that is reactive to sites of unsaturation.

Other examples of surface treatments include charged atmosphere treatments such as corona discharge treatment or plasma treatment. Electron beam treatment is also useful.

Interlayer adhesion may also be enhanced using an agent such as an aliphatic di- or
15 polyamine. The amine can be of any molecular weight that, when used, will result in an improvement in the adhesive bond strength between the layers of the multi-layer article. A particularly useful polyamine is polyallylamine having a molecular weight greater than about 1,000, as measured by gel permeation chromatography. An example of a useful commercially
20 available polyamine is polyallyl amine having a molecular weight of about 3,000 available from Nitto Boseki Co., Ltd.

The amine may be incorporated into one or more of the layers of the multi-layer article prior to forming the article using conventional means such as melt-mixing. Alternatively, the amine may be applied to a surface of one or more of the layers using conventional coating
25 methods such as spray coating, curtain coating, immersion coating, dip coating, and the like.

The invention will now be described further by way of the following examples.

EXAMPLES

The following examples describe the preparation of various multi-layer articles featuring a fluoroplastic layer bonded to an elastomer layer. In each example, the elastomer
30 was a fluoroelastomer prepared by combining the following ingredients: 100 parts Dyneon

FE-5830Q fluoroelastomer (commercially available from Dyneon LLC, St. Paul, MN); 13 parts N-762 carbon black (commercially available from Cabot Corp., Alpharetta, GA); 6 parts calcium hydroxide HP (commercially available from C.P. Hall, Chicago, IL); 3 parts magnesium oxide (commercially available from Morton International, Danvers, MA, under the designation "ElastomagTM 170"); and 6 parts calcium oxide HP (commercially available from C.P. Hall, Danvers, MA). The composition was extruded to form the fluoroelastomer in the shape of a tube having an outer diameter of 12 mm and a wall thickness of 0.33 mm.

Example 1

A cross-head die equipped with a polytetrafluoroethylene (PTFE) sleeve was used to coat a molten fluoroplastic composition onto the surface of the fluoroelastomer tube. The fluoroplastic was a TFE-HFP-VDF terpolymer featuring 76 wt.% TFE, 11 wt.% HFP, and 13 wt.% VDF. The fluoroplastic had a melt flow index of 7 and a melting point of 233°C. The PTFE sleeve prevented heating of the fluoroelastomer surface prior to application of the fluoroplastic.

Following application of the fluoroplastic composition, the resulting multi-layer article was passed through a 15.2 cm long tubular heater set at 220°C (the surface temperature of the fluoroplastic was 140°C) to heat the article prior to cooling. Once cooled, the article was cut into smaller samples that were then placed on a steel mandrel and thermally cured at a temperature of 160°C and a pressure of 0.4 MPa for 60 minutes using steam in an autoclave. Following cure, the samples were removed from the autoclave and cooled to room temperature.

The peel adhesion of the cured samples was evaluated by making a cut in each sample to separate a 7 mm wide strip of the fluoroplastic outer layer from the fluoroelastomer core in order to provide a tab for adhesion testing. The thickness of the fluoroplastic layer was 0.3 mm. An Instron® Model 1125 tester, available from Instron Corp., set at a 100 mm/min. crosshead speed was used as the test device. Peel strength between the fluoroplastic and fluoroelastomer layers was measured in accordance with ASTM D 1876 (T-Peel Test) with the exception that the peel angle was 90 degrees. The results of two samples were averaged. The average value is reported in Table 1.

Example 2

The procedure of Example 1 was followed except that the fluoroplastic was a TFE-HFP-VDF terpolymer commercially available from Dyneon LLC, St. Paul, MN under the designation "THV-500". The results of the peel adhesion test are reported in Table 1.

5 Comparative Example C-1

The procedure of Example 1 was followed except that the PTFE sleeve was not used. The results of the peel adhesion test are reported in Table 1.

Comparative Example C-2

10 The procedure of Example 1 was followed except that the heater was not used. The results of the peel adhesion test are reported in Table 1.

Comparative Example C-3

The procedure of Example 1 was followed except that neither the PTFE sleeve nor the heater was used. The results of the peel adhesion test are reported in Table 1.

TABLE 1

Example Number	Sleeve	Heater	Peel Strength (N/cm)
1	Yes	Yes	25.6
2	Yes	Yes	25.8
C-1	No	Yes	14.1
C-2	Yes	No	8.0
C-3	No	No	4.9

15

The results shown in Table 1 demonstrate that thermally insulating the curable elastomer layer prior to application of the fluoroplastic composition, in combination with heating the fluoroplastic layer following application of the fluoroplastic composition to the curable elastomer layer, results in multi-layer articles with enhanced interlayer adhesion upon

20 cure, even in the absence of separate adhesion-promoting measures.

In another set of examples, a multi-layer tube includes an inner layer of a fluoroelastomer, an intermediate layer of a fluorothermoplastic barrier layer, and an outer layer of an elastomer or rubber or thermoplastic elastomer.

Example 3

In Example 3, a cross-head die with a PTFE sleeve was used to coat THV-500 onto an extruded fluoroelastomer tube, which has an outer diameter of 16 mm with 1 mm thick wall. The sleeve prevented heating of the surface of the fluoroelastomer. The fluoroelastomer compound formulation for making the tube is shown in Table 2.

TABLE 2

Ingredients (supplier)	FKM comp
	phr*
Dyneon FE-5830Q (FKM) (Dyneon)	100
N-990 (carbon black) (Cancarb)	12
Vulcan XC072 (conductive carbon black) (Cabot)	10
Calcium hydroxide HP (C.P. Hall)	5
Elastomag™ 170 (magnesium oxide) (Morton International)	3
Calcium oxide HP (C.P. Hall)	6
Dibutyl sebacate (DBS) (Aldrich Chemical)	5

*All amounts referred to are in parts per 100 parts rubber by weight, abbreviated "phr."

Following application of the fluoroplastic composition, the resulting multi-layer article was passed through a 15.2 cm long tubular heater set at 220°C (the surface temperature of the fluoroplastic was 140°C) to heat the article prior to cooling. The fluoroplastic coated fluoroelastomer tube was cooled and then the tube was covered with ethylene-epichlorohydrin rubber (ECO) rubber, which had a wall thickness of 2 mm. The article was cut into curing samples. The samples were cured at 143°C and 0.28 MPa for 30 minutes by steam in an autoclave with a steel mandrel and then cured at 154°C and 0.41 MPa for 30 minute. Following the cure, the samples were removed from the autoclave and cooled to room temperature.

The peel adhesion of the cured samples was evaluated by making a cut to separate a 25.4 mm wide strip of the fluoroplastic layer from the fluoroelastomer and ECO layer from the fluoroplastic in order to provide tabs to test the adhesion between the layers via a peel test. The thickness of fluoroplastic layer was 0.3 mm. An Instron® Model 1125 tester, available

from Instron Corp., set at a 100 mm/mm crosshead speed was used as the test device. Peel strength or adhesion was measured on the two strips in accordance with ASTM D 1876 (T-Peel Test). The results of the two samples were averaged the test results are summarized in Table 3.

5 Example 4

In Example 4, the sample was prepared and tested as in Example 3 except that the first curing condition was 146°C and 0.3 MPa for 30 minutes. The test result is summarized in Table 3.

Comparative Example C-4

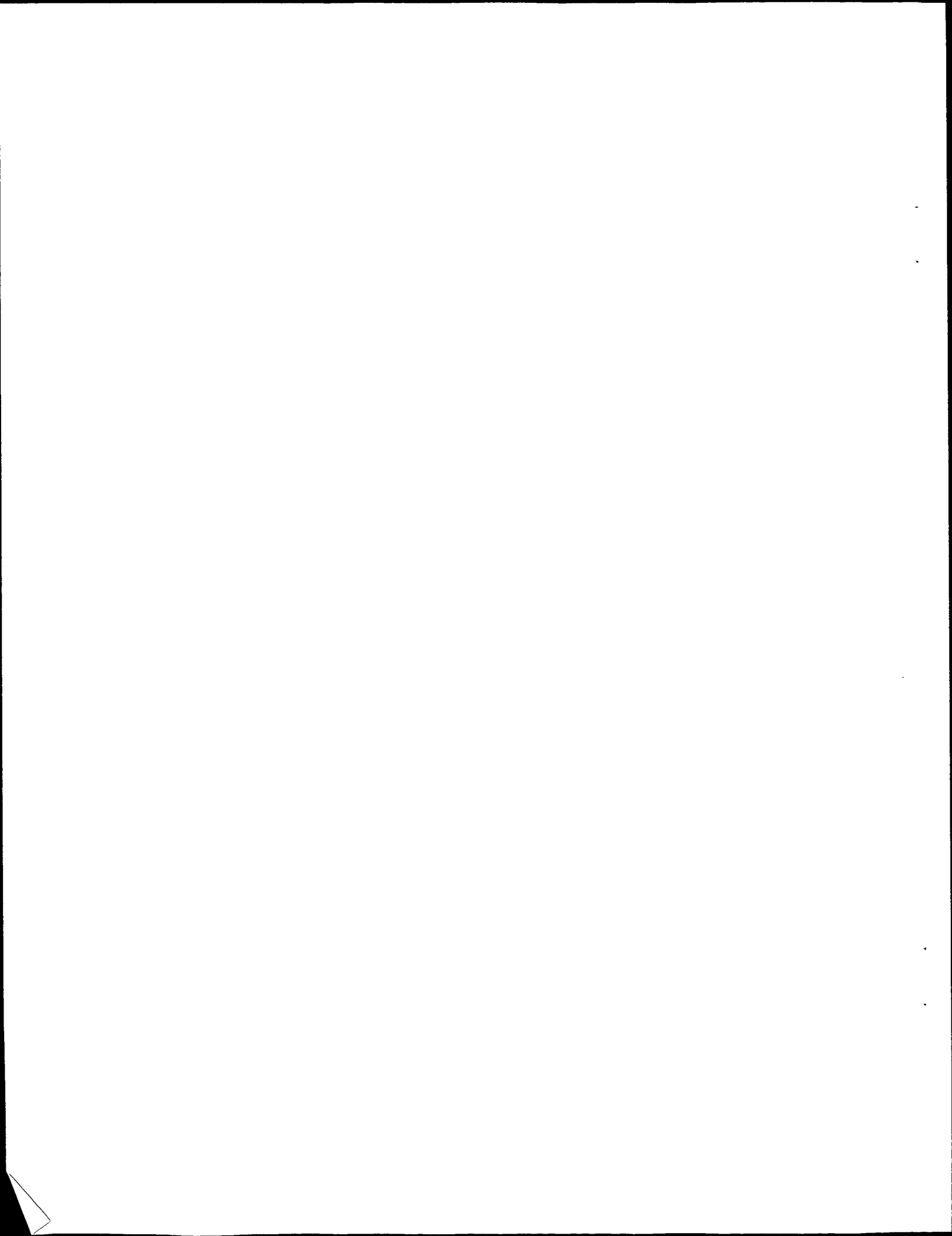
10 In Comparative Example C-1, the sample was prepared and tested as in Example 3 except that the sample was cured at 143°C and 0.28 MPa for 60 minutes without applying the second curing condition. The test result is summarized in Table 3.

Comparative Example C-5

15 In Comparative Example C-5, the sample was prepared and tested as in Example 3 except that the sample was cured at 154°C and 0.41 MPa for 30 minutes without applying the second curing condition. The test result is summarized in Table 3.

TABLE 3

Example	Curing Condition						Peel strength (N/cm)	
	1 st cure			2 nd cure				
	Pressure (MPa)	Temp. (°C)	Time (min)	Pressure (MPa)	Temp. (°C)	Time (min)	FKM/THV	THV/ECO
3	0.28	143	30	0.41	154	30	42	38
4	0.30	146	30	0.41	154	30	38	33
C-4	0.28	143	60	--	--	--	30	0.5
C-5	0.41	154	30	--	--	--	0.3	40



The data in Table 3 demonstrate that a step curing process provides substantially improved peel strength of both FKM/THV layer and THV/ECO layer in an article compared to the peel strength in an article prepared without step curing.

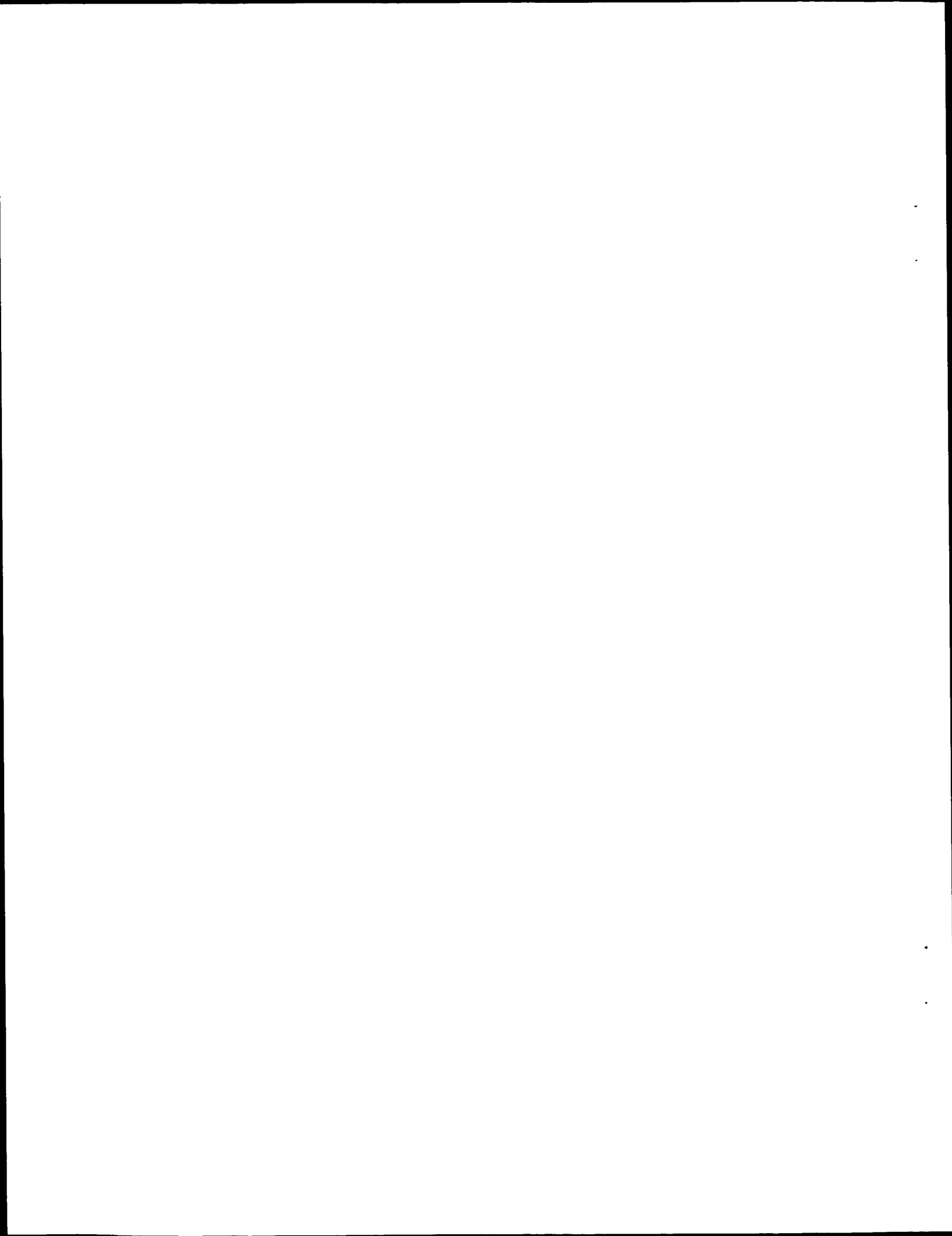
A number of embodiments of the invention have been described. Nevertheless, it will be understood that various modifications may be made without departing from the spirit and scope of the invention. Accordingly, other embodiments are within the scope of the following claims.

For example, although the process shown in Fig. 1 illustrates the preparation of a multi-layer article in the form of a tube, other shapes may be prepared as well. Also, while Fig. 1 illustrates the use of extruders to prepare the curable elastomer layer and fluoroplastic layers, other polymer processing techniques may be used. For example, the curable elastomer and fluoroplastic compositions can be prepared in the form of sheets and then laminated together, so long as measures are taken to thermally insulate the curable elastomer prior to application of the fluoroplastic. In addition, although Fig. 1 illustrates the use of a tubular heater for radiantly heating the fluoroplastic layer, other heating methods could be used. For example, in the case of fluoroplastic layers containing, e.g., metal particles, induction heating could be used.

WHAT IS CLAIMED IS:

1. A process for preparing a multi-layer article comprising:
 - (a) providing a precursor article comprising a curable elastomer layer, said article having an exposed surface available for application of a fluoroplastic layer;
 - 5 (b) thermally insulating said curable elastomer layer prior to application of said fluoroplastic layer;
 - (c) applying a fluoroplastic composition comprising interpolymerized vinylidene fluoride units onto said exposed surface of said precursor article to form a fluoroplastic layer;
 - (d) heating said fluoroplastic layer; and
 - 10 (e) curing said curable elastomer layer to form a multi-layer article comprising a fluoroplastic layer and an elastomer layer.
2. A process according to claim 1 comprising applying said fluoroplastic composition in molten form.
3. A process according to claim 2 comprising applying said fluoroplastic composition by
15 extrusion coating said fluoroplastic composition through a crosshead die onto said exposed surface of said precursor article.
4. A process according to claim 3 wherein said die comprises a die body that receives said fluoroplastic composition, an upstream opening for receiving said precursor article, a downstream opening, and a sleeve located at least partially within said upstream opening of
20 said die that receives said precursor article and thermally insulates said curable elastomer layer prior to application of said fluoroplastic composition.
5. A process according to claim 1 further comprising cooling said multi-layer article subsequent to heating said fluoroplastic layer.
6. A process according to claim 1 comprising thermally curing said curable elastomer
25 layer.

7. A process according to claim 1 comprising curing said curable elastomer layer subsequent to heating said fluoroplastic layer.
8. A process according to claim 1 comprising providing said precursor article by extruding a curable elastomer composition through a die to form said precursor article.
- 5 9. A process according to claim 1 wherein said curable elastomer layer has an exposed surface available for application of said fluoroplastic composition and said fluoroplastic composition is applied directly to said exposed surface of said curable elastomer layer.
10. A process according to claim 1 wherein said elastomer comprises a fluoroelastomer.
11. A process according to claim 1 wherein said elastomer comprises a non-fluorinated
10 elastomer.
12. A process according to claim 1 wherein said fluoroplastic has a melting temperature ranging from about 100 to about 330°C.
13. A process according to claim 1 wherein said fluoroplastic has a melting temperature ranging from about 150 to about 270°C.
- 15 14. A process according to claim 1 wherein said fluoroplastic comprises interpolymers derived from tetrafluoroethylene, vinylidene fluoride, and a monomer selected from the group consisting of hexafluoropropylene, perfluorinated alkoxy vinyl ethers, perfluorinated alkyl vinyl ethers, olefins, and combinations thereof.
15. A process according to claim 14 wherein the amount of said vinylidene fluoride units
20 is at least 3% by weight but less than 20% by weight.
16. A process according to claim 14 wherein the amount of said vinylidene fluoride units is between 10 and 15% by weight.



17. A process according to claim 1 further comprising bonding a polymer layer to said fluoroplastic layer to form a multi-layer article comprising said fluoroplastic layer interposed between said elastomer layer and said polymer layer.

18. A process according to claim 17 comprising bonding said polymer layer directly to
5 said fluoroplastic layer.

19. A process according to claim 17 wherein said polymer comprises an elastomer.

20. A process according to claim 19 wherein said elastomer comprises a nitrile rubber.

21. A process according to claim 1 further comprising placing a polymer layer on said fluoroplastic layer prior to curing.

10 22. A process according to claim 21 wherein curing includes a first stage at a first temperature and a second stage at a second temperature, the first temperature being lower than the second temperature.

23. A process according to claim 22 wherein said polymer comprises an elastomer.

24. A process according to claim 1 wherein said multi-layer article is in the form of a tube.

25. A process according to claim 1 wherein the adhesion between said fluoroplastic layer and said elastomer layer is at least 15 N/cm.

26. A process according to claim 22 wherein the adhesion between said fluoroplastic layer
5 and said polymer layer is at least 15 N/cm.

27. A process according to claim 1 further comprising cooling said curable elastomer layer prior to application of said fluoroplastic composition.

28. A process for preparing a multi-layer article comprising:

(a) providing a precursor article comprising a curable elastomer layer, said curable
10 elastomer layer having an exposed surface available for application of a fluoroplastic layer;
(b) extrusion coating a molten fluoroplastic composition comprising interpolymers of vinylidene fluoride units through a crosshead die onto said exposed surface of said curable elastomer layer to form a fluoroplastic layer,

said die comprising a die body that receives said molten fluoroplastic composition, an
15 upstream opening for receiving said precursor article, a downstream opening, and a sleeve located at least partially within said upstream opening of said die that receives said precursor article and thermally insulates said curable elastomer layer prior to application of said fluoroplastic composition;

(c) heating said fluoroplastic layer; and

20 (d) thermally curing said curable elastomer layer subsequent to heating said fluoroplastic layer to form a multi-layer article comprising a fluoroplastic layer and an elastomer layer.

29. A process for preparing a multi-layer article comprising:

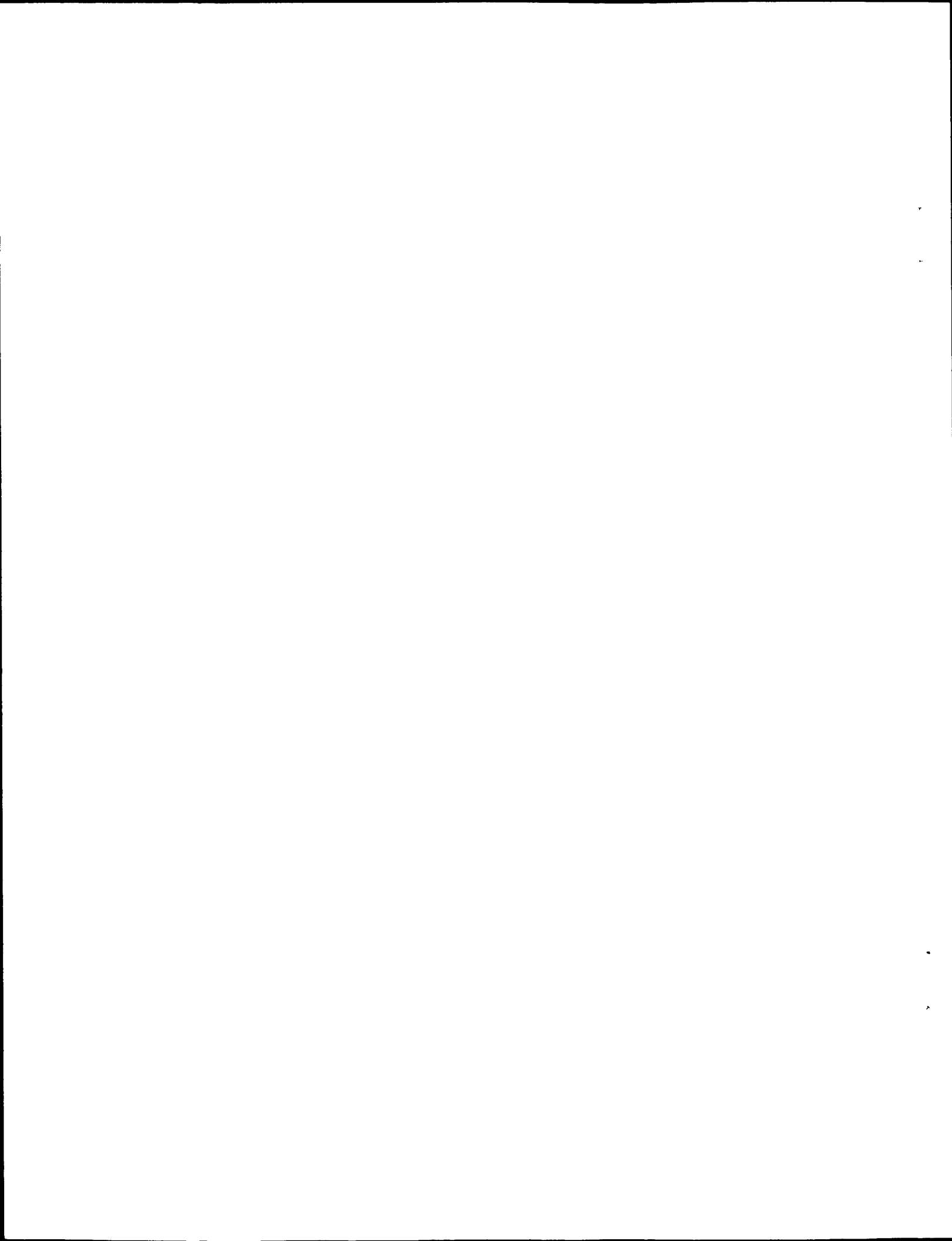
(a) providing a precursor article comprising a curable elastomer layer, said curable
25 elastomer layer having an exposed surface available for application of a fluoroplastic layer;

(b) extrusion coating a molten fluoroplastic composition comprising interpolymerized vinylidene fluoride units through a crosshead die onto said exposed surface of said curable elastomer layer to form a fluoroplastic layer,

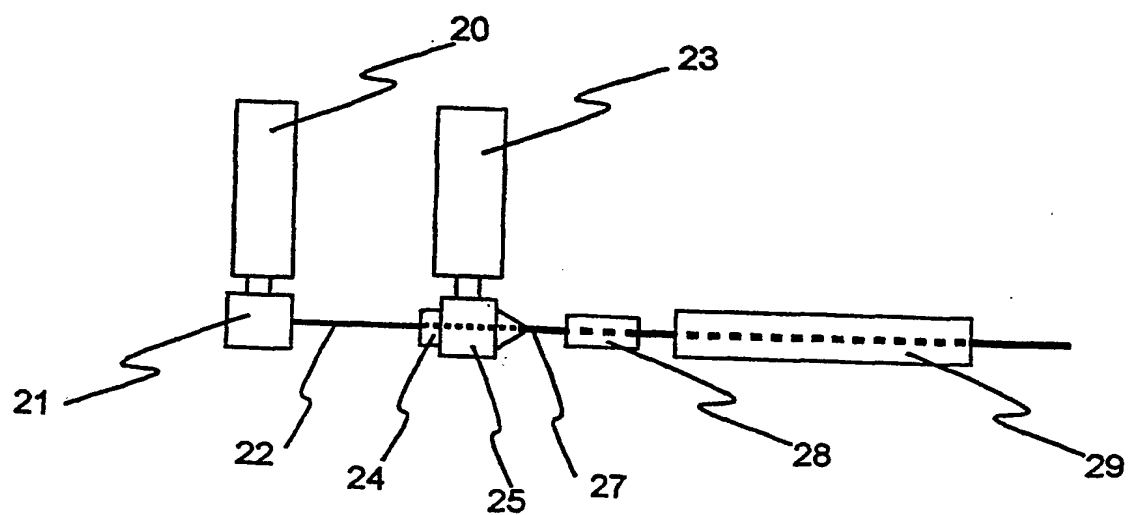
said die comprising a die body that receives said molten fluoroplastic composition, an
5 upstream opening for receiving said precursor article, a downstream opening, and a sleeve located at least partially within said upstream opening of said die that receives said precursor article and thermally insulates said curable elastomer layer prior to application of said fluoroplastic composition;

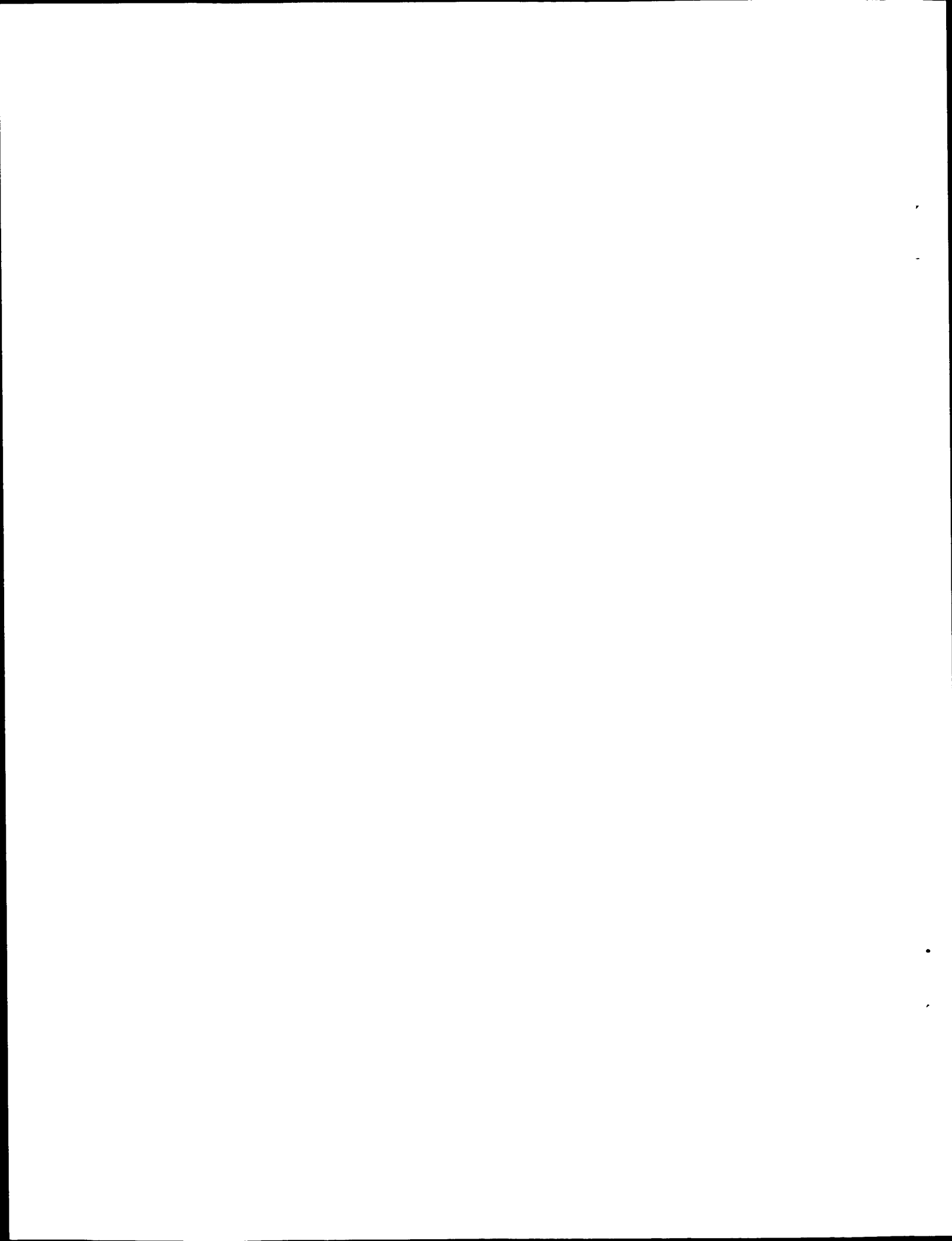
(c) placing a polymer layer on said fluoroplastic layer; and

10 (d) thermally curing said elastomer layer and polymer layer in a first stage at a first temperature and a second stage at a second temperature, the first temperature being lower than the second temperature, to form a multi-layer article comprising a fluoroplastic layer, an elastomer layer, and a polymer layer.



1/1

**Fig. 1**



INTERNATIONAL SEARCH REPORT

Int ☐ national Application No

PCT/US 01/24867

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 B29C47/06 B32B1/08 B32B27/08 F16L9/12 F16L11/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 B29C B32B F16L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 641 445 A (DUKES GLENN V ET AL) 24 June 1997 (1997-06-24) column 1, line 15 -column 2, line 23 column 2, line 52 -column 3, line 55 column 5, line 63 -column 6, line 1 column 6, line 46 - line 56; claims 1,2,10,20-22	1,2, 8-10, 12-19, 24,28
X A	column 6, line 56 -column 7, line 56 claims 19,23-25	21,23 29
A	WO 96 00657 A (DUKES GLENN V ;BROWDER WILLIAM TROY (US); CADILLAC RUBBER & PLASTI) 11 January 1996 (1996-01-11) claims 1,7,11,14,18,19	1,2, 8-10, 12-19, 21,23, 24,28,29

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☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

* Special categories of cited documents:

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Date of the actual completion of the international search

5 October 2001

Date of mailing of the international search report

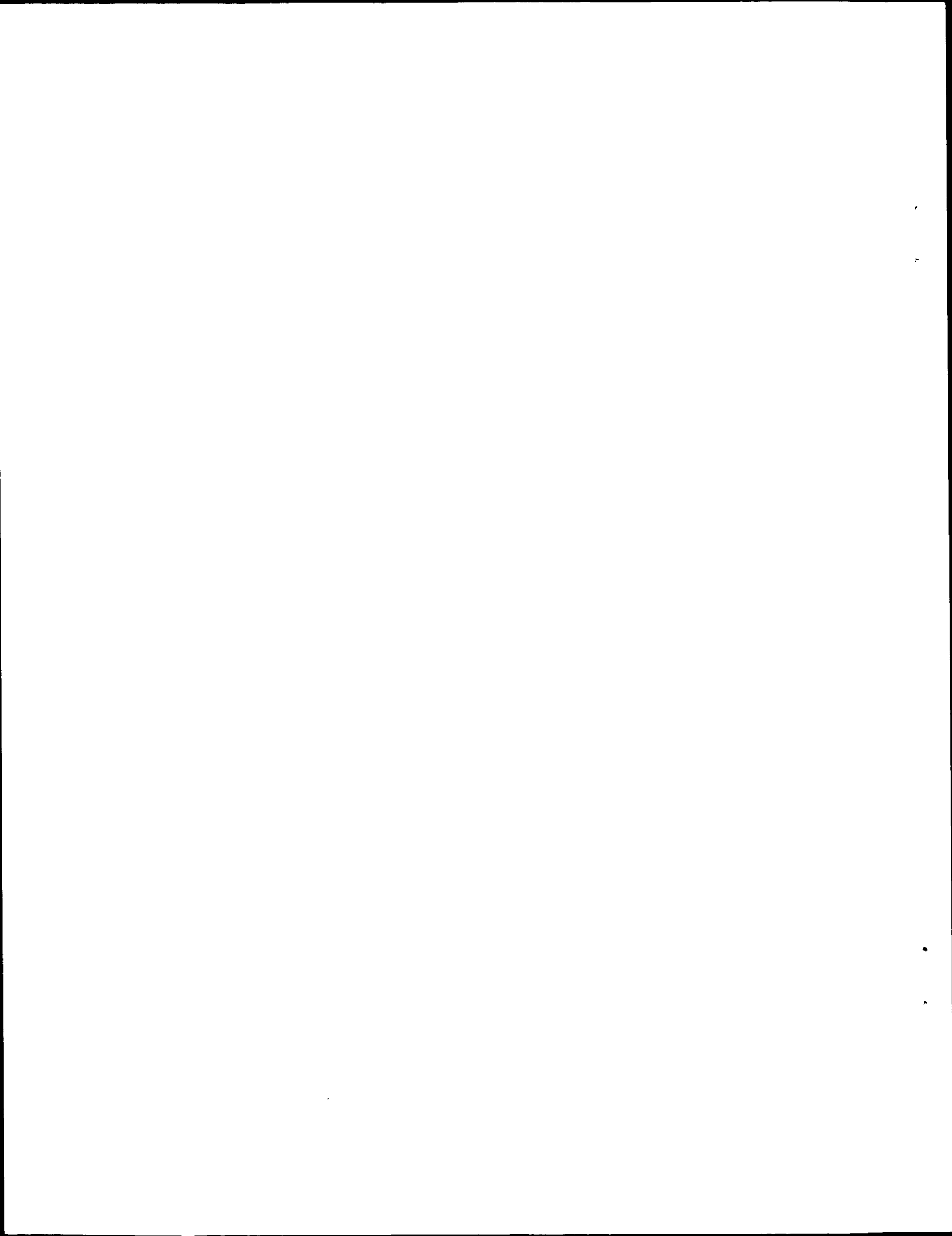
12/10/2001

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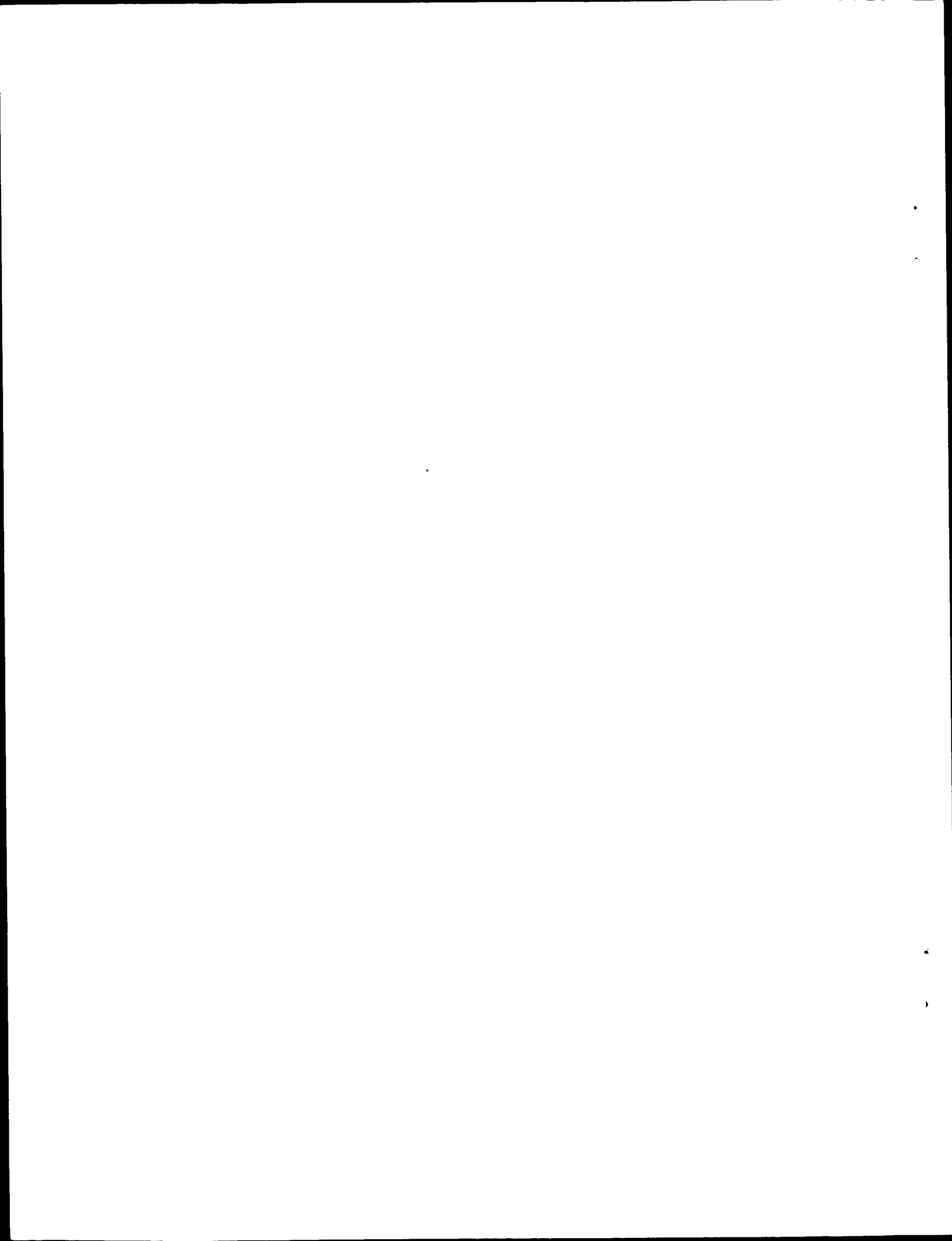


INTERNATIONAL SEARCH REPORT

Int 1al Application No
PCT/US 01/24867

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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A	US 4 895 744 A (BRIGGS MILTON ET AL) 23 January 1990 (1990-01-23) column 1, line 64 -column 3, line 17; figure 4 column 11, line 22 - line 40 ---	21-23,29
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Information on patent family members

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NOTIFICATION CONCERNING REPRESENTATION

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JAMES V. LILLY
OFFICE OF INTELLECTUAL PROPERTY
COUNSEL
POST OFFICE BOX 33427
SAINT PAUL, MN 55133 3427

Applicant's or agent's file reference <div style="text-align: center;">55791WO007</div>	Date of mailing <i>(day/month/year)</i> <div style="text-align: center; font-size: 1.2em;">31 OCT 2001</div>
International application No. <div style="text-align: center;">PCT/US01/24867</div>	International filing date <i>(day/month/year)</i> <div style="text-align: center;">08 AUG 01</div>
Applicant 3M INNOVATIVE PROPERTIES COMPANY	

1. This receiving Office hereby gives notice of the receipt of a document containing:

- ☒ a power of attorney
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2. This notification, together with a copy of the document indicated above, is sent to the addressee in its capacity as:

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- ☐ the International Bureau, which is requested to record a change in the person of the agent or common representative under Rule 92*bis*.1(a)(ii).

Name and mailing address of the receiving Office Assistant Commissioner for Patent Box PCT Washington, D.C. 20231 Attn:RO/US Facsimile No. 703-305-3230	Authorized officer Eric Simms Telephone No. 703-305-3761
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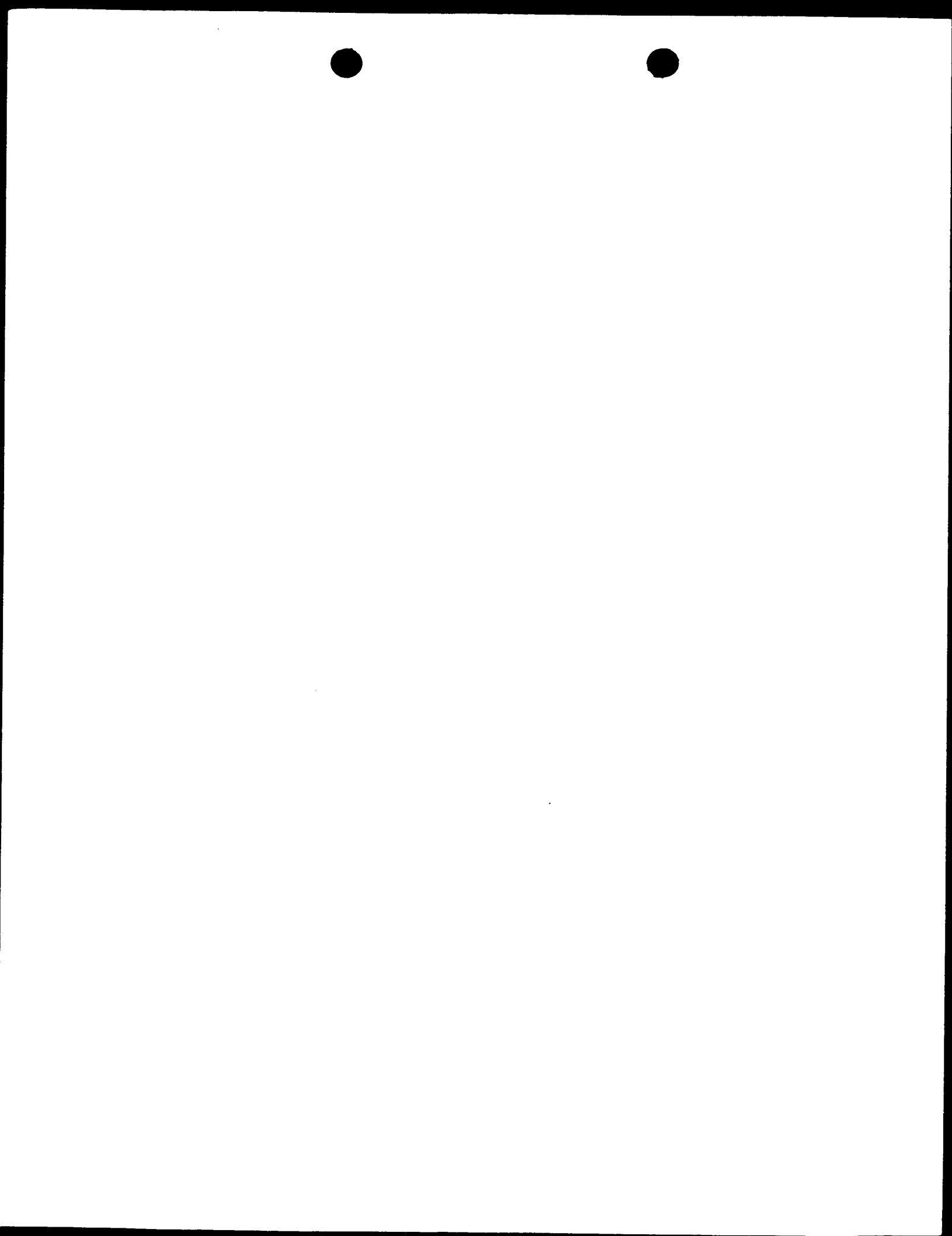
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PCT POWER OF ATTORNEY

55791WO007

Printed on 03.08.2001 09:17:40 AM

0-1	PCT Power of Attorney (for an international application filed under the Patent Cooperation Treaty) (PCT Rule 90.4)	
0-1-1	Prepared using	PCT-EASY Version 2.92 (updated 01.03.2001)
1	The undersigned applicant(s)	FUKUSHI, Tatsuo; KOLB, Robert E.; HOFF, Craig R.; WELLNER, Steven J.; MOLNAR, Attila
1-1-1	hereby appoints (appoint) the following person	LILLY, James V.; GRISWOLD, Gary L.; BATES, Carolyn A.; BOEDER, Jennie G.; CHERNIVEC, Gerald F.; LITTLE, Douglas B.; SPRAGUE, Robert W. Office of Intellectual Property Counsel Post Office Box 33427 Saint Paul, MN 55133-3427 United States of America
1-2	as	agent
1-3	to represent the undersigned before	all the competent International Authorities
1-4	In connection with the International application identified below:	
1-4-1	Title of the invention	PROCESS FOR PREPARING A MULTI-LAYER ARTICLE HAVING A FLUOROPLASTIC LAYER AND AN ELASTOMER LAYER
1-4-2	Applicant's or agent's file reference	55791WO007
1-4-3	International application number (if already available)	
1-4-4	filed with the following Office as receiving Office	United States Patent and Trademark Office (USPTO) (RO/US)
1-5	and to make or receive payments on behalf of the undersigned.	
2-2	Signature of applicant	<i>Tatsuo Fukushi</i> 8-7-2001
2-2-1	Name	FUKUSHI, Tatsuo
2-3	Signature of applicant	<i>Robert E. Kolb</i> 8/7/2001
2-3-1	Name	KOLB, Robert E.
2-4	Signature of applicant	<i>Craig Hoff</i> 8-7-01
2-4-1	Name	HOFF, Craig R.



ANNEX A TO FORM PCT/RO/106

International application No.

PCT/US01/24867

The receiving Office has found the following defects in the international application as filed:

1. As to **signature*** of the international application (Rules 4.15 and 90.4), the request:

- a. ☐ is not signed.
- b. ☒ is not signed by all applicants.
- c. ☐ is not accompanied by the statement referred to in the check list in Box No. VIII of the request explaining the lack of the signature of an applicant for the designation of the United States of America.
- d. ☐ is signed by what appears to be an agent/common representative but
 - ☐ the international application is not accompanied by a power of attorney appointing him.
 - ☐ the power of attorney accompanying the international application was not signed by all the applicants.
- e. ☐ other (*specify*):

* All applicants must sign, including inventors if they are also applicants (e.g. where the United States of America is designated).

2. As to indications concerning the **applicant**, the request (Rules 4.4 and 4.5):

- a. ☐ does not properly indicate the applicant's name (*specify*):
- b. ☐ does not indicate the applicant's address.
- c. ☐ does not properly indicate the applicant's address (*specify*):
- d. ☐ does not indicate the applicant's nationality.
- e. ☐ does not indicate the applicant's residence.
- f. ☐ other (*specify*):

3. As to the **language** of certain elements of the international application, other than the description and claims (Rules 12.1(c) and 26.3ter(a) and (c)):

- a. ☐ the **request** is not in a language which is both a language accepted by this receiving Office and a language of publication, which is (are):
- b. ☐ the **text matter of the drawings** is not in the language in which the international application is to be published, which is:
- c. ☐ the **abstract** is not in the language in which the international application is to be published, which is:

4. The **title** of the invention:

- a. ☐ is not indicated in Box No. I of the request (Rule 4.1(a)).
- b. ☐ is not indicated at the top of the first sheet of the description (Rule 5.1(a)).
- c. ☐ as appearing in Box No. I of the request is not identical with the title heading the description (Rule 5.1(a)).

5. As to the **abstract** (Rule 8):

- ☐ the international application does not contain an abstract.



PCT APPLICATION NUMBER: 01/24867

JAMES V. LILLY
OFFICE OF INTELLECTUAL PROPERTY COUNSEL
POST OFFICE BOX 33427
SAINT PAUL MN 55133-3427



**** THIS IS NOT AN ORDER ****

Certification Order Information

Date: 15 AUG 01

The following order will be electronically submitted to the CERTIFICATION DIVISION, OFFICE OF PUBLIC RECORDS, after the Record Copy has been sent to the INTERNATIONAL BUREAU. Please verify the following information and correct any errors in the PALM system prior to shipping the Record Copy.

US Serial Number: 09644731

US Filing Date: 23 AUG 00

○ Applicant: FUKUSHI
KOLB

Title: PROCESS FOR PREPARING A MULTI-LAYER ARTICLE HAVING
A FLUOROPLASTIC LAYER AND AN ELASTOMER LAYER

Atty Docket Num: 08772-009001

PCT Application: PCT/US01/24867

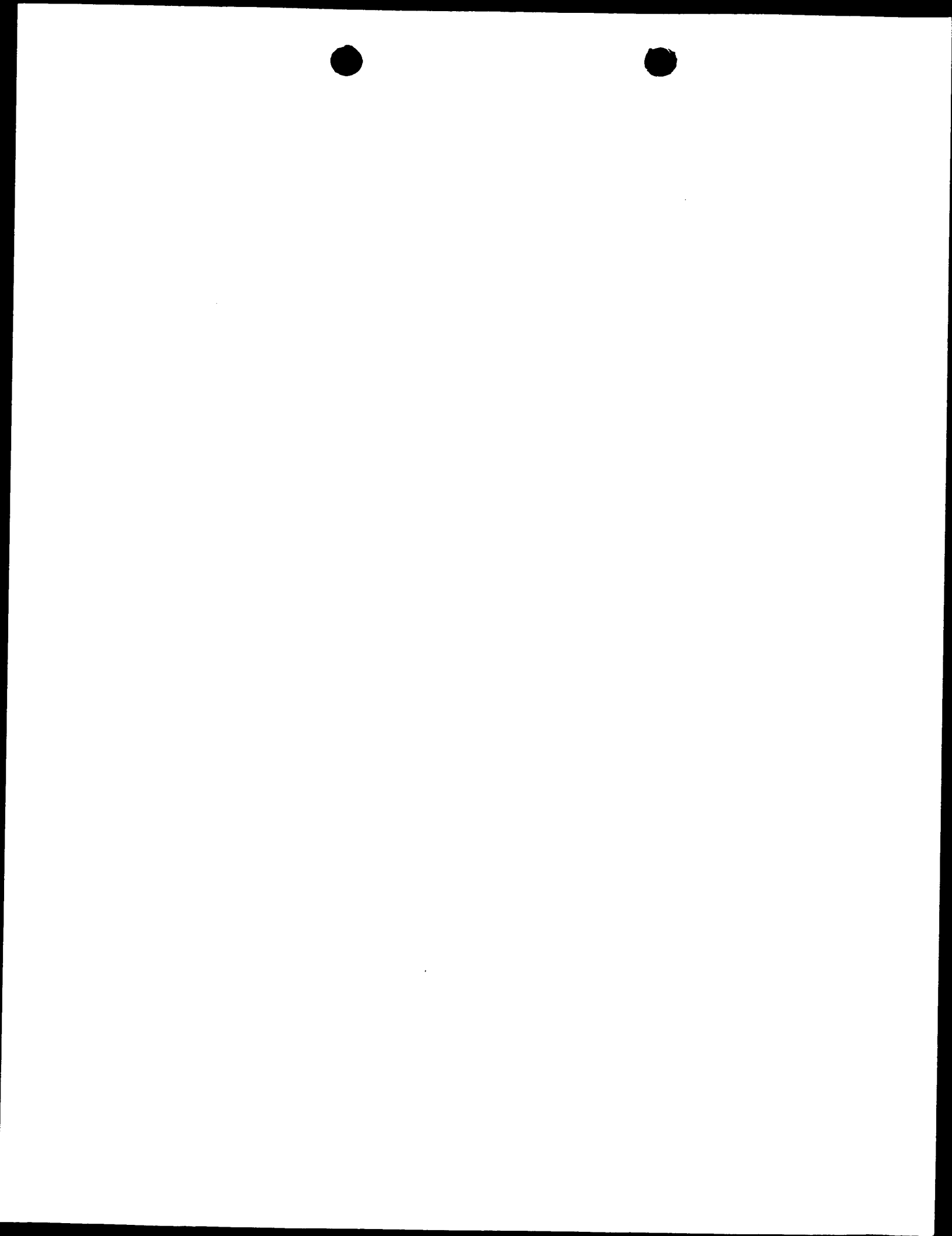
PCT Filing Date: 08 AUG 01

● Applicant : 3M INNOVATIVE PROPERTIES COMPANY

Title: PROCESS FOR PREPARING A MULTI-LAYER ARTICLE HAVING
A FLUOROPLASTIC LAYER AND AN ELASTOMER LAYER

Atty Docket Num: 55791W0007

**** THIS IS NOT AN ORDER ****



UNITED STATES RECEIVING OFFICE(RO/US) FEE CODING AND RECORDING SHEET

☐ ADD'L
SHEETS

IDENTIFICATION OF THE INTERNATIONAL APPLICATION

INTERNATIONAL APPLICATION NUMBER
PCT/US01/24867

INTERNATIONAL FILING DATE
08 AUG 01

APPLICANT (Name)
3M INNOVATIVE PROPERTIES COMPANY

PAYMENTS

REFUNDS

Payment on Filing			Deposit Account	Deposit Account	To Deposit Account	To Deposit Account
Deposit Account	13	3723	DATE:	DATE:	DATE:	DATE:
<input type="checkbox"/> CASH/CHECK			<input type="checkbox"/> CASH/CHECK	<input type="checkbox"/> CASH/CHECK	<input type="checkbox"/> BY CHECK	<input type="checkbox"/> BY CHECK
150	240.00					
151						
153						
800						
810	265.00					
801						
802	846.00					
899	492.00					
566	15.00	Total Paid:	Total Paid:	Total Paid:	Total Refunded:	Total Refunded:
		\$1858.00				
States Included for 892:			892:	892:		
States Included for 893:			893:	893:		

CALCULATED FEE AMOUNT = \$1858.00

AMOUNT OF DIFFERENCE = + \$0.00

RO/US Authorization

RO/US Authorization

RO/US Authorization

RO/US Authorization

RO/US Authorization



PATENT COOPERATION TREATY

From the RECEIVING OFFICE

PCT

To:

JAMES V. LILLY
OFFICE OF INTELLECTUAL PROPERTY COUNSEL
POST OFFICE BOX 33427
SAINT PAUL MN 55133-3427

COMMUNICATION IN CASES FOR WHICH
NO OTHER FORM IS APPLICABLE

Applicant's or agent's file reference 55791W0007	Date of mailing (day/month/year)
International application No. PCT/US01/24867	REPLY DUE See paragraph 1 below International filing date (day/month/year) 08 AUG 01
Applicant 3M INNOVATIVE PROPERTIES COMPANY	

1. ☐ REPLY DUE within **ONE MONTH** from the above date of mailing
- ☐ NO REPLY DUE, however, see below _____
- ☐ IMPORTANT COMMUNICATION
- ☐ INFORMATION ONLY

2. COMMUNICATION:

Name and mailing address of the receiving Office Assistant Commissioner for Patents Box PCT Washington, D.C. 20231 Facsimile No.	Authorized officer Telephone No.
--	---

PATENT COOPERATION TREATY

From the RECEIVING OFFICE

PCT

To:

JAMES V. LILLY
OFFICE OF INTELLECTUAL PROPERTY COUNSEL
POST OFFICE BOX 33427
SAINT PAUL MN 55133-3427

NOTIFICATION REGARDING CERTAIN CORRECTIONS MADE *EX OFFICIO*

(PCT Administrative Instructions, Section 327)

<p>Date of mailing (day/month/year)</p>	
<p>Applicant's or agent's file reference 55791W0007</p>	<p>REPLY DUE NONE However, see paragraph 3 below</p>
<p>International application No. PCT/US01/24867</p>	<p>International filing date (day/month/year) 08 AUG 01</p>
<p>Applicant 3M INNOVATIVE PROPERTIES COMPANY</p>	

1. The applicant is hereby notified that this receiving Office has corrected formal defects in the international application *ex officio*, as shown on the attached copy of:

- ☐ the request, sheet No.: _____
- ☐ the description, sheet No.: _____
- ☐ the claims, sheet No.: _____
- ☐ the drawings, sheet No.: _____
- ☐ other (*specify*): _____

2. If the applicant agrees with these corrections, no further action is required in this regard.

3. In case of disagreement with these corrections, the applicant should promptly inform this receiving Office accordingly.

<p>Name and mailing address of the receiving Office Assistant Commissioner for Patents Box PCT Washington, D.C. 20231 Facsimile No.</p>	<p>Authorized officer</p> <p style="text-align: center;">Attn: RO/US</p> <p>Telephone No.</p>
---	---



PATENT COOPERATION TREATY

From the INTERNATIONAL SEARCHING AUTHORITY

PCT

To:

JAMES V. LILLY
OFFICE OF INTELLECTUAL PROPERTY COUNSEL
POST OFFICE BOX 33427
SAINT PAUL MN 55133-3427

NOTIFICATION OF RECEIPT
OF SEARCH COPY

(PCT Rule 25.1)

To: JAMES V. LILLY OFFICE OF INTELLECTUAL PROPERTY COUNSEL POST OFFICE BOX 33427 SAINT PAUL MN 55133-3427		PCT	
Date of mailing (day/month/year)		IMPORTANT NOTIFICATION	
Applicant's or agent's file reference 55791W0007			
International application No. PCT/US01/24867	International filing date (day/month/year) 08 AUG 01	Priority date (day/month/year) 23 AUG 00	
Applicant 3M INNOVATIVE PROPERTIES COMPANY			

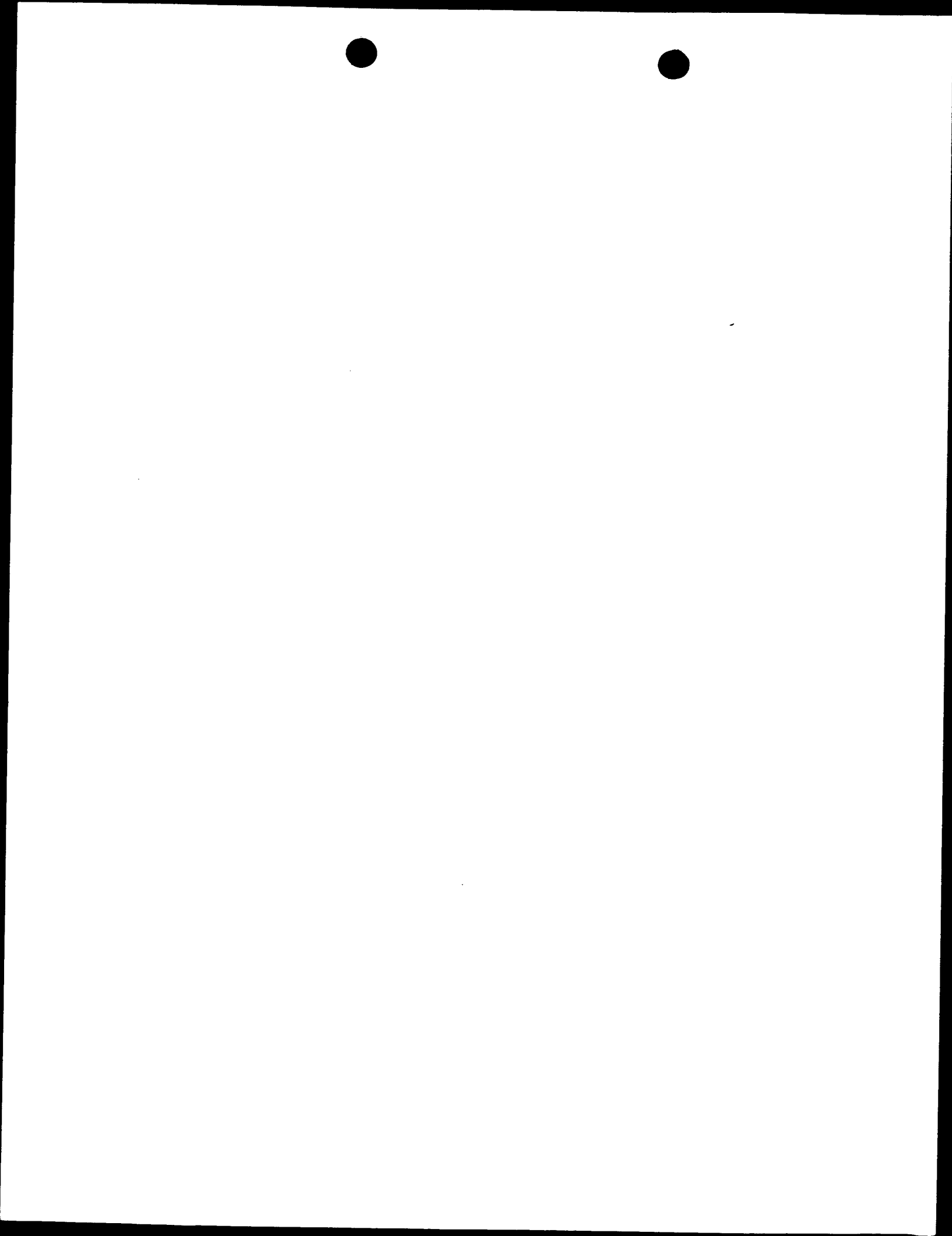
1. **Where the International Searching Authority and the receiving Office are not the same Office:**
 The applicant is hereby notified that the search copy of the international application was received by this International Searching Authority on the date indicated below.

Where the International Searching Authority and the receiving Office are the same Office:
 The applicant is hereby notified that the search copy of the international application was received on the date indicated below.

_____ (date of receipt)

2. **Time limit for establishment of international search report**
 The applicant is informed that the time limit for establishing the international search report is 3 months from the date of receipt indicated above or 9 months from the priority date, whichever time limit expires later.
3. A copy of this notification has been sent to the International Bureau and, where the first sentence of paragraph 1 applies, to the receiving Office.

Name and mailing address of the ISA/US Assistant Commissioner for Patents Box PCT Washington, D.C. 20231 Facsimile No.	Authorized officer Telephone No.
--	---



PATENT COOPERATION TREATY

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

PCT

To:

JAMES V. LILLY
OFFICE OF INTELLECTUAL PROPERTY COUNSEL
POST OFFICE BOX 33427
SAINT PAUL MN 55133-3427

NOTIFICATION OF RECEIPT OF DEMAND BY COMPETENT INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

(PCT Rule 59.3(e) and 61.1(b), first sentence
and Administrative Instructions, Section 601(a))

Date of mailing
(day/month/year)

Applicant's or agent's file reference
55791W0007

IMPORTANT NOTIFICATION

International application No.
PCT/US01/24867

International filing date (day/month/year)
08 AUG 01

Priority date (day/month/year)
23 AUG 00

Applicant

3M INNOVATIVE PROPERTIES COMPANY

1. The applicant is hereby **notified** that this International Preliminary Examining Authority considers the following date as the date of receipt of the demand for international preliminary examination of the international application:

2. That date of receipt is:

- ☐ the actual date of receipt of the demand by this Authority (Rule 61.1(b)).
- ☐ the actual date of receipt of the demand on behalf of this Authority (Rule 59.3(e)).
- ☐ the date on which this Authority has, in response to the invitation to correct defects in the demand (Form PCT/IPEA/404), received the required corrections.

3. ☐ **ATTENTION:** That date of receipt is **AFTER** the expiration of 19 months from the priority date. Consequently, the election(s) made in the demand does (do) not have the effect of postponing the entry into the national phase until 30 months from the priority date (or later in some Offices) (Article 39(1)). Therefore, the acts for entry into the national phase must be performed within 20 months from the priority date (or later in some Offices) (Article 22). For details, see the *PCT Applicant's Guide*, Volume II.

- ☐ (If applicable) This notification confirms the information given by telephone, facsimile transmission or in person on:

4. Only where paragraph 3 applies, a copy of this notification has been sent to the International Bureau.

Name and mailing address of the IPEA/US
Assistant Commissioner for Patents
Box PCT
Washington, D.C. 20231
Facsimile No.

Attn: IPEA/US

Authorized officer

Telephone No.



TO

JAMES V. LILLY
OFFICE OF INTELLECTUAL PROPERTY COUNSEL
POST OFFICE BOX 33427
SAINT PAUL MN 55133-3427

UNITED STATES DESIGNATED/ELECTED
OFFICE (DO/EO/US)

NOTIFICATION OF STATUS OF
REQUIREMENTS UNDER 35 U.S.C.371

DATE OF MAILING

FILE REFERENCE

55791W0007

IDENTIFICATION OF INTERNATIONAL APPLICATION

International Application Number	International Filing Date	Priority Date Claimed
PCT/US01/24867	08 AUG 01	23 AUG 00

Applicant for DO/EO/US

3M INNOVATIVE PROPERTIES COMPANY

NOTIFICATION

The applicant is hereby advised that the U.S. Patent and Trademark Office in its capacity as ☐ Designated Office ☐ Elected Office has received the following items as of the date of mailing indicated above.

1. ☐ U.S. National fee [35 U.S.C.371 (c) (1)]
 2. ☐ Oath of declaration [35 U.S.C.371 (c) (4)]
 3. ☐ Copy of International application as filed [35 U.S.C.371 (c) (2)]
 4. ☐ Translation of Application [35 U.S.C.371 (c) (2)]
 5. ☐ Amendments under PCT Article 19 [35 U.S.C.371 (c) (3)]
 6. ☐ Translation of PCT Article 19 Amendments [35 U.S.C.371 (c) (3)]
 7. ☐ Search Report or Declaration under PCT Article 17(2) [35 U.S.C.371 (a)]
 8. ☐ International Preliminary Examination Report and its Annexes, if any, under PCT Article 36(3) (a) [35 U.S.C.371 (a)]
 9. ☐ Translation of Annexes to the International Preliminary Examination Report under PCT Article 36(3) (b) [35 U.S.C.371 (c) (5)]
 10. ☐ Other items received:
☐ Assignment Document ☐ Prior Art Statement ☐ Preliminary Amendment
- A. ☐ Requirements for U.S. National processing have been met. Processing will commence
☐ at the expiration of the applicable time limit under either
☐ PCT Article 22 [35 U.S.C.371 (b)] or
☐ PCT Article 39 [35 U.S.C.371 (b)]
☐ on the date indicated below under the provisions of 35 U.S.C.371 (f)

U.S. NATIONAL SERIAL#

DATE UNDER 35 U.S.C.102(e)

DATE OF COMMENCEMENT OF
NATIONAL PROCESSING

All correspondence submitted after the date of commencement of U.S. National processing indicated above should refer to the U.S. National Serial Number and the appropriate U.S. National processing organization or Officer.

- B. ☐ As the above identified application has been accepted for U.S. National processing under the provisions of 35 U.S.C.371 (f) before expiration of the applicable time limit under ☐ PCT Article 22 ☐ PCT Article 39, applicant is reminded that
- ☐ Amendments under PCT Article 19 and/or
 - ☐ the International Preliminary Examination Report and its Annexes, if any, under PCT Article 36(3) (a), and (b)
- and any translation thereof, if applicable, must be submitted to the Patent and Trademark Office as soon as they are available.



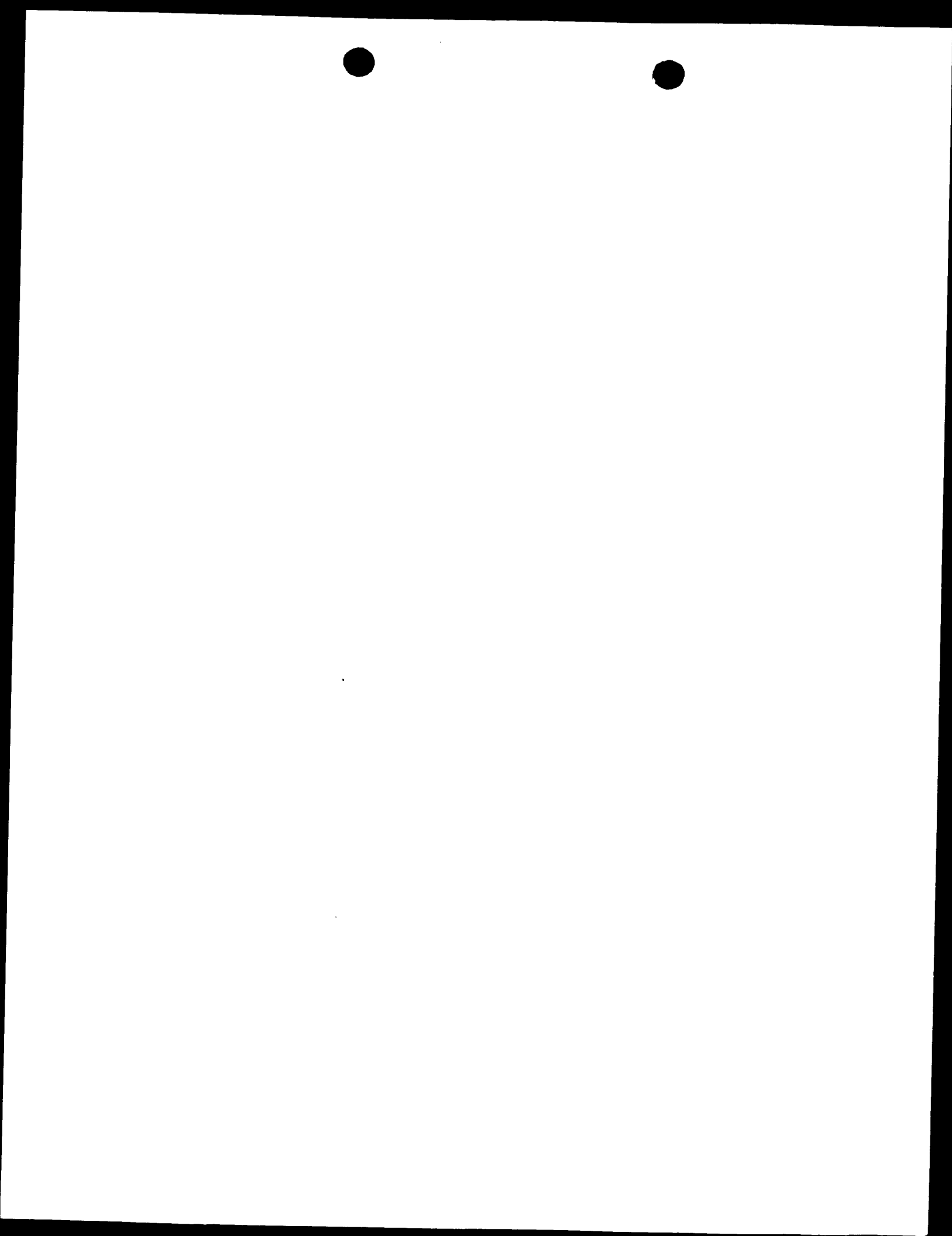
PCT (ANNEX - FEE CALCULATION SHEET)

Original (for SUBMISSION) - printed on 03.08.2001 09:20:43 AM

(This sheet is not part of and does not count as a sheet of the international application)

0	For receiving Office use only		PCT/US01 / 24867	
0-1	International Application No.			
0-2	Date stamp of the receiving Office			
0-4	Form - PCT/RO/101 (Annex)			
0-4-1	PCT Fee Calculation Sheet Prepared using		PCT-EASY Version 2.92 (updated 01.03.2001)	
0-9	Applicant's or agent's file reference		55791WO007	
2	Applicant		3M INNOVATIVE PROPERTIES COMPANY, et al.	
12	Calculation of prescribed fees		fee amount/multiplier	total amounts (USD)
12-1	Transmittal fee	T	⇒	240
12-2	Search fee	S	⇒	846
12-3	International fee			
	Basic fee			
	(first 30 sheets)	b1	382	
12-4	Remaining sheets		0	
12-5	Additional amount	(X)	9	
12-6	Total additional amount	b2	0	
12-7	b1 + b2 =	B	382	
12-8	Designation fees			
	Number of designations contained in international application		89	
12-9	Number of designation fees payable (maximum 6)		6	
12-10	Amount of designation fee	(X)	82	
12-11	Total designation fees	D	492	
12-12	PCT-EASY fee reduction	R	-117	
12-13	Total International fee (B+D-R)	I	⇒	757
12-14	Fee for priority document			
	Number of priority documents requested		1	
12-15	Fee per document	(X)	15	
12-16	Total priority document fee	P	⇒	15
12-17	TOTAL FEES PAYABLE (T+S+I+P)		⇒	1,858
12-19	Mode of payment		authorization to charge deposit account	
12-20	Deposit account instructions			
	The receiving Office:		United States Patent and Trademark Office (USPTO) (RO/US)	
12-20-1	Authorization to charge the total fees indicated above.		✓	

Duplicate



PCT (ANNEX - FEE CALCULATION SHEET)

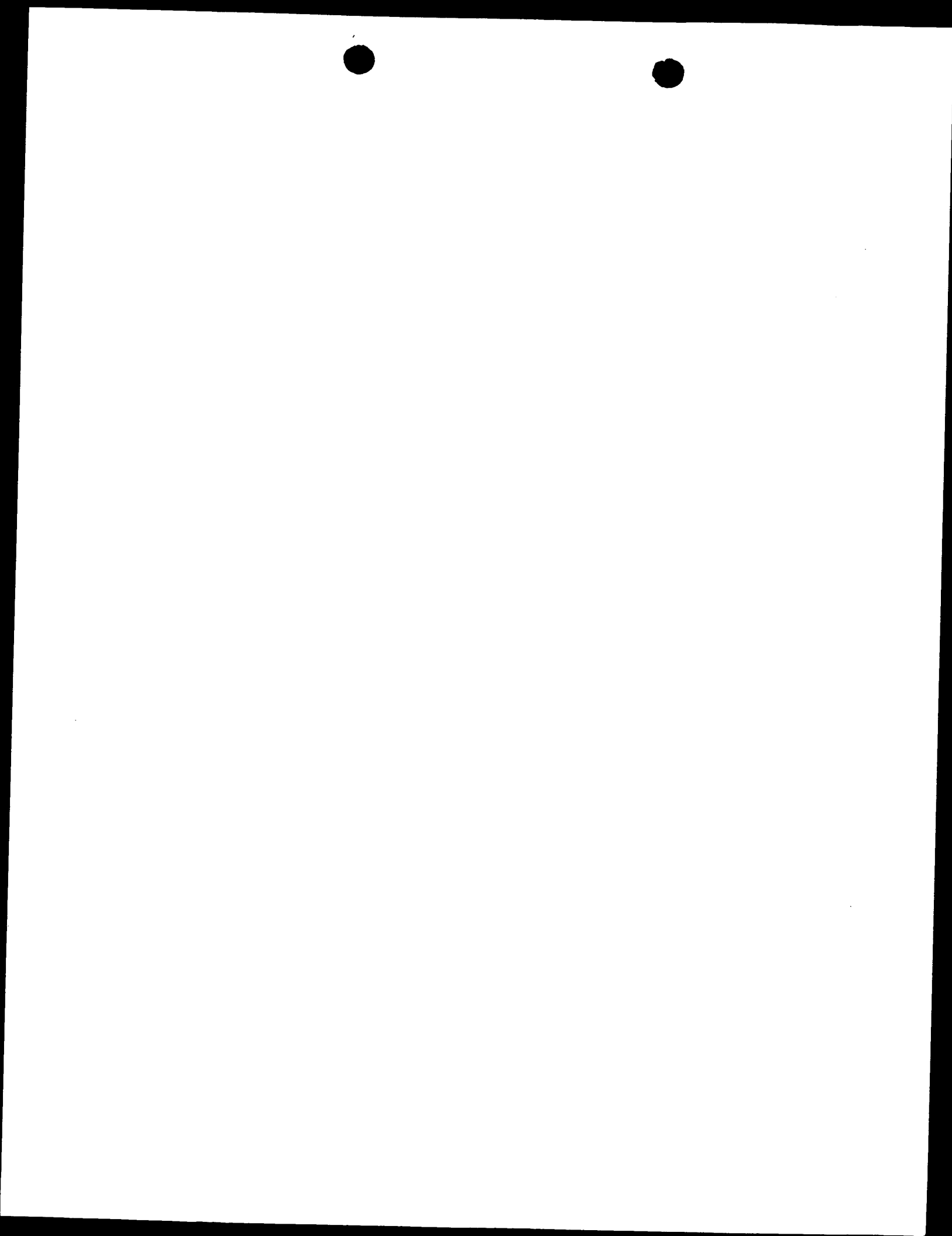
Original (for SUBMISSION) - printed on 03.08.2001 09:20:43 AM

12-20-2	Authorization to charge any deficiency or credit any overpayment in the total fees indicated above.	✓
12-21	Deposit account No.	13-3723
12-22	Date	03 August 2001 (03.08.2001)
12-23	Name and signature	Douglas B. Little, Assistant Chief Intellectual Property Counsel <i>Douglas B. Little</i>

VALIDATION LOG AND REMARKS

13-2-3	Validation messages Names	Green? Applicant 3.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Applicant 4.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Applicant 5.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Agent 1.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Agent 2.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Agent 3.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Agent 4.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.

Duplicate



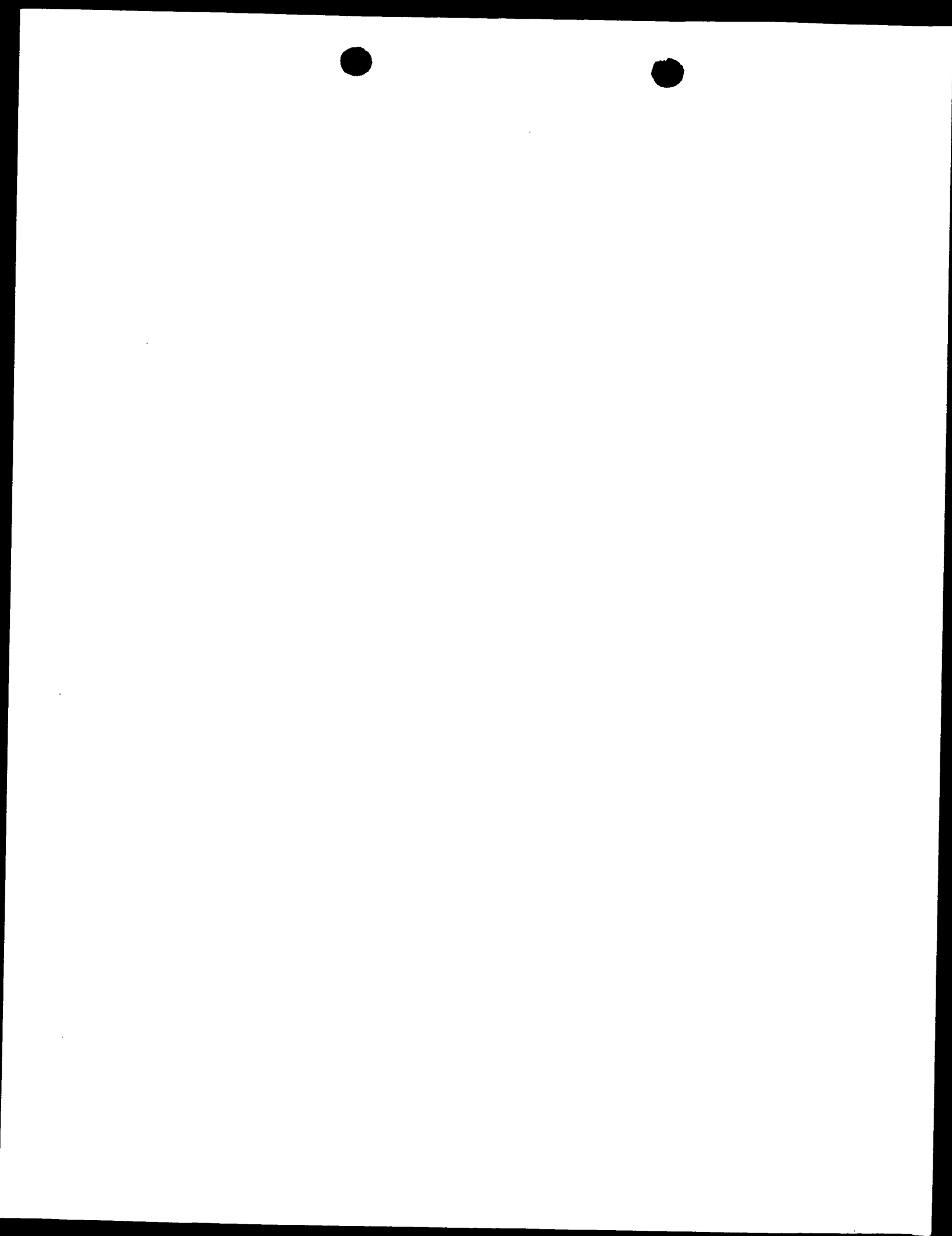
HOMECOPY

1/6

55791WO007

PCT REQUESTOriginal (for **SUBMISSION**) - printed on 03.08.2001 09:20:43 AM

0	For receiving Office use only	PCT/US01/24867
0-1	International Application No.	
0-2	International Filing Date	08 AUG 2001 (08.08.01)
0-3	Name of receiving Office and "PCT International Application"	PCT/US
0-4	Form - PCT/RO/101 PCT Request	
0-4-1	Prepared using	PCT-EASY Version 2.92 (updated 01.03.2001)
0-5	Petition The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty	
0-6	Receiving Office (specified by the applicant)	United States Patent and Trademark Office (USPTO) (RO/US)
0-7	Applicant's or agent's file reference	55791WO007
I	Title of invention	PROCESS FOR PREPARING A MULTI-LAYER ARTICLE HAVING A FLUOROPLASTIC LAYER AND AN ELASTOMER LAYER
II	Applicant	
II-1	This person is:	applicant only
II-2	Applicant for	all designated States except US
II-4	Name	3M INNOVATIVE PROPERTIES COMPANY
II-5	Address:	3M Center Post Office Box 33427 Saint Paul, MN 55133-3427 United States of America
II-6	State of nationality	US
II-7	State of residence	US
II-8	Telephone No.	(651) 733-1500
II-9	Facsimile No.	(651) 736-7586
III-1	Applicant and/or inventor	
III-1-1	This person is:	applicant and inventor
III-1-2	Applicant for	US only
III-1-4	Name (LAST, First)	FUKUSHI, Tatsuo
III-1-5	Address:	1882 Bowsens Lane Woodbury, MN 55125 United States of America
III-1-6	State of nationality	JP
III-1-7	State of residence	US



PCT REQUEST

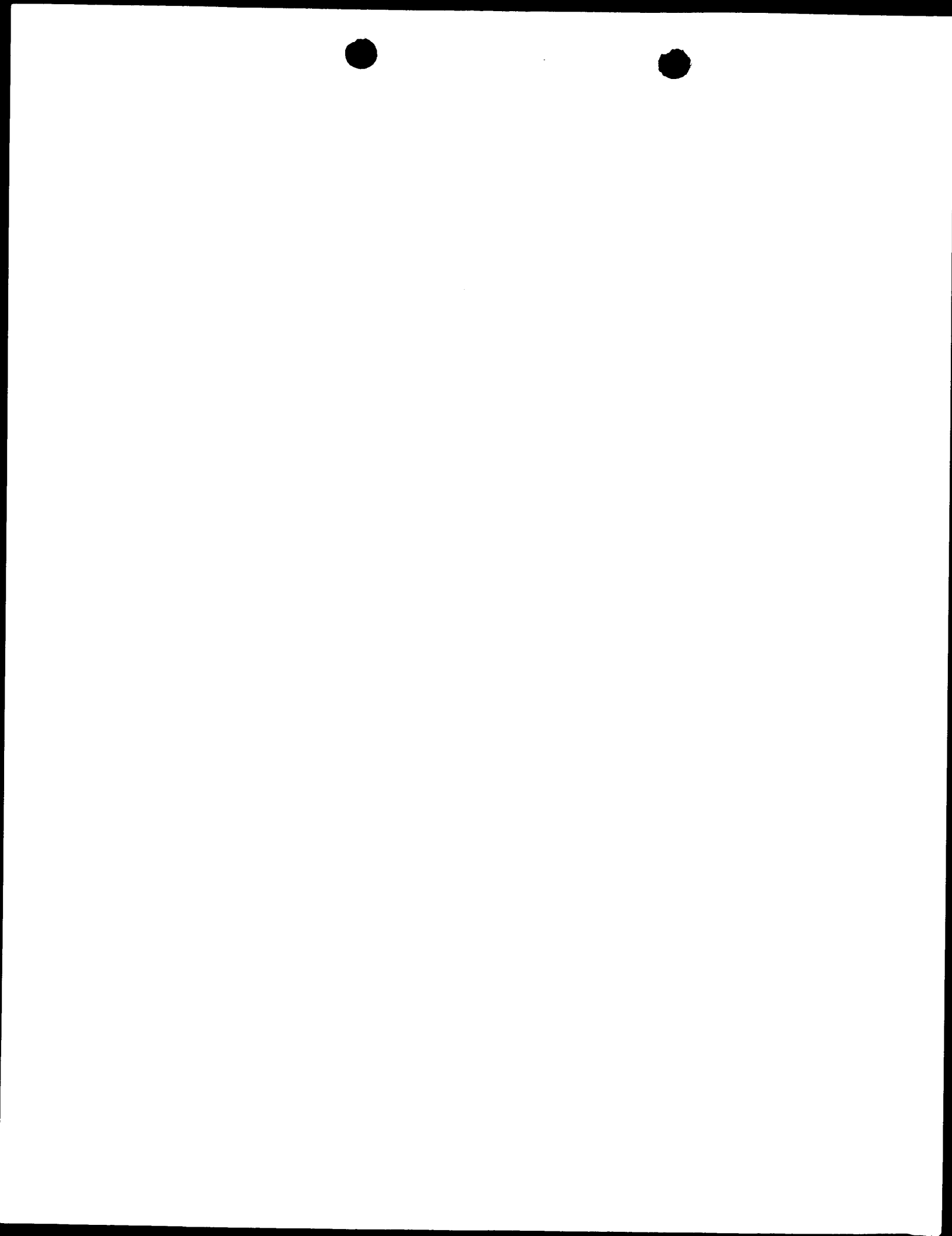
Original (for SUBMISSION) - printed on 03.08.2001 09:20:43 AM

III-2	Applicant and/or inventor	
III-2-1	This person is:	applicant and inventor
III-2-2	Applicant for	US only
III-2-4	Name (LAST, First)	KOLB, Robert E.
III-2-5	Address:	3486 Trading Post Trail South Afton, MN 55128 United States of America
III-2-6	State of nationality	US
III-2-7	State of residence	US
III-3	Applicant and/or inventor	
III-3-1	This person is:	applicant and inventor
III-3-2	Applicant for	US only
III-3-4	Name (LAST, First)	HOFF, Craig R.
III-3-5	Address:	8040 120th Avenue Champlin, MN 55316 United States of America
III-3-6	State of nationality	US
III-3-7	State of residence	US
III-4	Applicant and/or inventor	
III-4-1	This person is:	applicant and inventor
III-4-2	Applicant for	US only
III-4-4	Name (LAST, First)	WELLNER, Steven J.
III-4-5	Address:	14676 Afton Boulevard South Afton, MN 55001 United States of America
III-4-6	State of nationality	US
III-4-7	State of residence	US
III-5	Applicant and/or inventor	
III-5-1	This person is:	applicant and inventor
III-5-2	Applicant for	US only
III-5-4	Name (LAST, First)	MOLNAR, Attila
III-5-5	Address:	4143 Primrose Path Vadnais Heights, MN 55127 United States of America
III-5-6	State of nationality	CA
III-5-7	State of residence	US

PCT REQUEST

Original (for SUBMISSION) - printed on 03.08.2001 09:20:43 AM

IV-1	Agent or common representative; or address for correspondence The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as:	agent
IV-1-1	Name (LAST, First)	LILLY, James V.
IV-1-2	Address:	Office of Intellectual Property Counsel Post Office Box 33427 Saint Paul, MN 55133-3427 United States of America
IV-1-3	Telephone No.	(651) 733-1543
IV-1-4	Facsimile No.	(651) 736-7586
IV-2	Additional agent(s)	additional agent(s) with same address as first named agent
IV-2-1	Name(s)	GRISWOLD, Gary L.; BATES, Carolyn A.; BOEDER, Jennie G.; CHERNIVEC, Gerald F.; LITTLE, Douglas B.; SPRAGUE, Robert W.
V	Designation of States	
V-1	Regional Patent (other kinds of protection or treatment, if any, are specified between parentheses after the designation(s) concerned)	AP: GH GM KE LS MW MZ SD SL SZ TZ UG ZW and any other State which is a Contracting State of the Harare Protocol and of the PCT EA: AM AZ BY KG KZ MD RU TJ TM and any other State which is a Contracting State of the Eurasian Patent Convention and of the PCT EP: AT BE CH&LI CY DE DK ES FI FR GB GR IE IT LU MC NL PT SE TR and any other State which is a Contracting State of the European Patent Convention and of the PCT OA: BF BJ CF CG CI CM GA GN GQ GW ML MR NE SN TD TG and any other State which is a member State of OAPI and a Contracting State of the PCT



PCT REQUEST

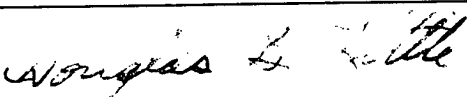
Original (for SUBMISSION) - printed on 03.08.2001 09:20:43 AM

V-2	National Patent (other kinds of protection or treatment, if any, are specified between parentheses after the designation(s) concerned)	AE AG AL AM AT (patent and utility model) AU AZ BA BB BG BR BY BZ CA CH&LI CN CO CR CU CZ (patent and utility model) DE (patent and utility model) DK (patent and utility model) DM DZ EC EE (patent and utility model) ES FI (patent and utility model) GB GD GE GH GM HR HU ID IL IN IS JP KE KG KP KR KZ LC LK LR LS LT LU LV MA MD MG MK MN MW MX MZ NO NZ PL PT RO RU SD SE SG SI SK (patent and utility model) SL TJ TM TR TT TZ UA UG US UZ VN YU ZA ZW
V-5	Precautionary Designation Statement In addition to the designations made under items V-1, V-2 and V-3, the applicant also makes under Rule 4.9(b) all designations which would be permitted under the PCT except any designation(s) of the State(s) indicated under item V-6 below. The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit.	
V-6	Exclusion(s) from precautionary designations	NONE
VI-1	Priority claim of earlier national application	
VI-1-1	Filing date	23 August 2000 (23.08.2000)
VI-1-2	Number	09/644,731
VI-1-3	Country	US
VI-2	Priority document request The receiving Office is requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) identified above as item(s):	VI-1
VII-1	International Searching Authority Chosen	European Patent Office (EPO) (ISA/EP)



PCT REQUEST

Original (for SUBMISSION) - printed on 03.08.2001 09:20:43 AM

VIII		Declarations		Number of declarations	
VIII-1	Declaration as to the identity of the inventor	-			
VIII-2	Declaration as to the applicant's entitlement, as at the international filing date, to apply for and be granted a patent	-			
VIII-3	Declaration as to the applicant's entitlement, as at the international filing date, to claim the priority of the earlier application	-			
VIII-4	Declaration of inventorship (only for the purposes of the designation of the United States of America)	-			
VIII-5	Declaration as to non-prejudicial disclosures or exceptions to lack of novelty	-			
IX		Check list		number of sheets	
IX-1	Request (including declaration sheets)	6			electronic file(s) attached
IX-2	Description	12			-
IX-3	Claims	5			-
IX-4	Abstract	1			EZABST00.TXT
IX-5	Drawings	1			-
IX-7	TOTAL	25			
		Accompanying items		paper document(s) attached	
IX-8	Fee calculation sheet		✓		electronic file(s) attached
IX-17	PCT-EASY diskette	-			Diskette
IX-18	Other (specified):	Record of Action Taken by Directors of 3M Innovative Properties			-
IX-18	Other (specified):	Transmittal Letter			-
IX-18	Other (specified):	Itemized Return Postcard			-
IX-19	Figure of the drawings which should accompany the abstract	1			
IX-20	Language of filing of the international application	English			
X-1	Signature of applicant, agent or common representative				
X-1-1	Name	3M INNOVATIVE PROPERTIES COMPANY			
X-1-2	Name of signatory	LITTLE, Douglas B.			
X-1-3	Capacity	Assistant Chief Intellectual Property Counsel			

FOR RECEIVING OFFICE USE ONLY

(08108101)

10-1	Date of actual receipt of the purported international application	513 Rec'd PCT/US 03 AUG 2001
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PCT REQUEST

Original (for SUBMISSION) - printed on 03.08.2001 09:20:43 AM

10-2	Drawings:	
10-2-1	Received	
10-2-2	Not received	
10-3	Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application	
10-4	Date of timely receipt of the required corrections under PCT Article 11(2)	
10-5	International Searching Authority	ISA/EP
10-6	Transmittal of search copy delayed until search fee is paid	

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11-1	Date of receipt of the record copy by the International Bureau	
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PCT POWER OF ATTORNEY

Printed on 03.08.2001 09:17:40 AM

0-1	PCT Power of Attorney (for an international application filed under the Patent Cooperation Treaty) (PCT Rule 90.4)	
0-1-1	Prepared using	PCT-EASY Version 2.92 (updated 01.03.2001)
1	The undersigned applicant(s)	FUKUSHI, Tatsuo; KOLB, Robert E.; HOFF, Craig R.; WELLNER, Steven J.; MOLNAR, Attila
1-1-1	hereby appoints (appoint) the following person	LILLY, James V.; GRISWOLD, Gary L.; BATES, Carolyn A.; BOEDER, Jennie G.; CHERNIVEC, Gerald F.; LITTLE, Douglas B.; SPRAGUE, Robert W. Office of Intellectual Property Counsel Post Office Box 33427 Saint Paul, MN 55133-3427 United States of America
1-2	as	agent
1-3	to represent the undersigned before	all the competent International Authorities
1-4	In connection with the international application identified below:	
1-4-1	Title of the invention	PROCESS FOR PREPARING A MULTI-LAYER ARTICLE HAVING A FLUOROPLASTIC LAYER AND AN ELASTOMER LAYER
1-4-2	Applicant's or agent's file reference	55791WO007
1-4-3	International application number (if already available)	
1-4-4	filed with the following Office as receiving Office	United States Patent and Trademark Office (USPTO) (RO/US)
1-5	and to make or receive payments on behalf of the undersigned.	
2-2	Signature of applicant	<i>Tatsuo Fukushi</i> 8-7-2001
2-2-1	Name	FUKUSHI, Tatsuo
2-3	Signature of applicant	<i>Robert E. Kolb</i> 8/7/2001
2-3-1	Name	KOLB, Robert E.
2-4	Signature of applicant	<i>Craig Hoff</i> 8-7-01
2-4-1	Name	HOFF, Craig R.

RO/US 01/24867
RO/US 20 SEP 2001

2/2

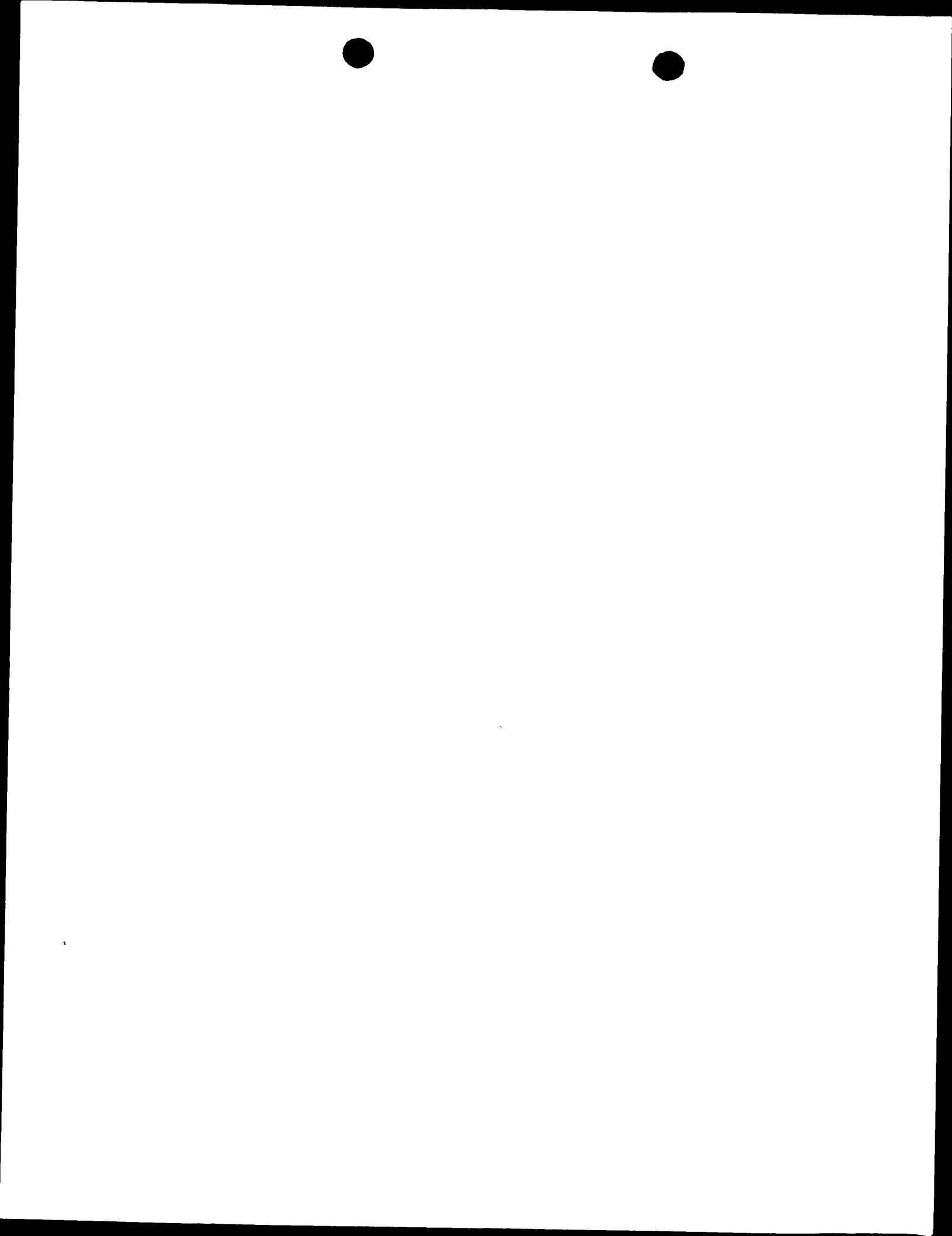
55791WO007

PCT POWER OF ATTORNEY

Printed on 03.08.2001 09:17:40 AM

2-5	Signature of applicant	<i>S. J. Wellner</i> 8/08/01
2-5-1	Name	WELLNER, Steven J.
2-6	Signature of applicant	<i>Attila Molnar</i> 8/7/01
2-6-1	Name	MOLNAR, Attila
3	Date	03 August 2001 (03.08.2001)

INSTITUTE SHEET (RULE 26)



**RECORD OF ACTION TAKEN BY DIRECTORS
OF 3M INNOVATIVE PROPERTIES COMPANY
April 23, 1999**

The undersigned, being all directors of 3M Innovative Properties Company, do hereby authorize in writing, without a meeting therefor, pursuant to §141(f) of Delaware General Corporation Law, the adoption of the following, effective the 23rd day of April, 1999:

**Authorization to Sign Documents
Relating to Intellectual Property**

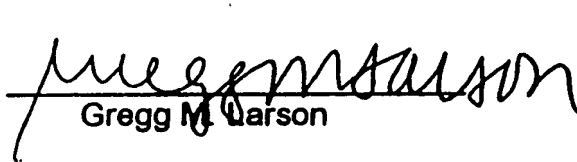
RESOLVED, That the Chief Intellectual Property Counsel, or an Associate Chief Intellectual Property Counsel, or an Assistant Chief Intellectual Property Counsel, or any of them, be and hereby are authorized to sign any and all documents or other materials in connection with (a) the filing, prosecution, maintenance, or ownership of any patent, trademark, or copyright application or any patent, trademark, or copyright anywhere in the world, or (b) litigation involving intellectual property including any patent, trademark, copyright, or trade secret matter anywhere in the world, on behalf of the Corporation.

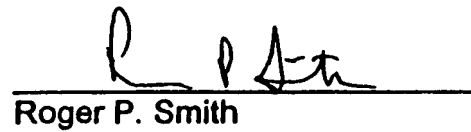
Election of Officers

The following persons are hereupon elected to the offices set forth after their names to serve until the next annual meeting or until their respective successors are duly elected and qualified:

Gary L. Griswold	-	President and Chief Intellectual Property Counsel
Paul F. Plotnik	-	Operations Manager, International Business Development
Roger P. Smith	-	Secretary
Carolyn A. Bates	-	Assistant Secretary
William J. Schmoll	-	Treasurer
Kimberly M. Torseth	-	Assistant Treasurer

IN WITNESS WHEREOF, The undersigned has subscribed their own names.


Gregg M. Larson


Roger P. Smith

There being no further business, the minutes are herewith closed.


Carolyn A. Bates
Assistant Secretary



PCT (ANNEX - FEE CALCULATION SHEET)

55791WO007

Original (for SUBMISSION) - printed on 03.08.2001 09:20:43 AM

(This sheet is not part of and does not count as a sheet of the international application)

0	For receiving Office use only		
0-1	International Application No.	PCT/US01/24867	
0-2	Date stamp of the receiving Office	08 AUG 2001 (08.08.01)	
0-4	Form - PCT/RO/101 (Annex) PCT Fee Calculation Sheet		
0-4-1	Prepared using	PCT-EASY Version 2.92 (updated 01.03.2001)	
0-9	Applicant's or agent's file reference	55791WO007	
2	Applicant	3M INNOVATIVE PROPERTIES COMPANY, et al.	
12	Calculation of prescribed fees	fee amount/multiplier	total amounts (USD)
12-1	Transmittal fee T	⇒	240 240.00
12-2	Search fee S	⇒	846 846.00
12-3	International fee		
	Basic fee		
	(first 30 sheets) b1	382	
12-4	Remaining sheets	0	
12-5	Additional amount (X)	9	
12-6	Total additional amount b2	0	
12-7	b1 + b2 = B	382	
12-8	Designation fees		
	Number of designations contained in international application	89	
12-9	Number of designation fees payable (maximum 6)	6	
12-10	Amount of designation fee (X)	82	
12-11	Total designation fees D	492	
12-12	PCT-EASY fee reduction R	-117	
12-13	Total International fee (B+D-R) I	⇒	757 757.00
12-14	Fee for priority document		
	Number of priority documents requested	1	
12-15	Fee per document (X)	15	
12-16	Total priority document fee P	⇒	15 15.00
12-17	TOTAL FEES PAYABLE (T+S+I+P)	⇒	1,858 1858.00
12-19	Mode of payment	authorization to charge deposit account	
12-20	Deposit account instructions		
	The receiving Office:	United States Patent and Trademark Office (USPTO) (RO/US)	
12-20-1	Authorization to charge the total fees indicated above.	✓	



PCT (ANNEX - FEE CALCULATION SHEET)

55791WO007

Original (for SUBMISSION) - printed on 03.08.2001 09:20:43 AM

12-20-2	Authorization to charge any deficiency or credit any overpayment in the total fees indicated above.	✓
12-21	Deposit account No.	13-3723
12-22	Date	03 August 2001 (03.08.2001)
12-23	Name and signature	Douglas B. Little, Assistant Chief Intellectual Property Counsel <i>Douglas B. Little</i>

VALIDATION LOG AND REMARKS

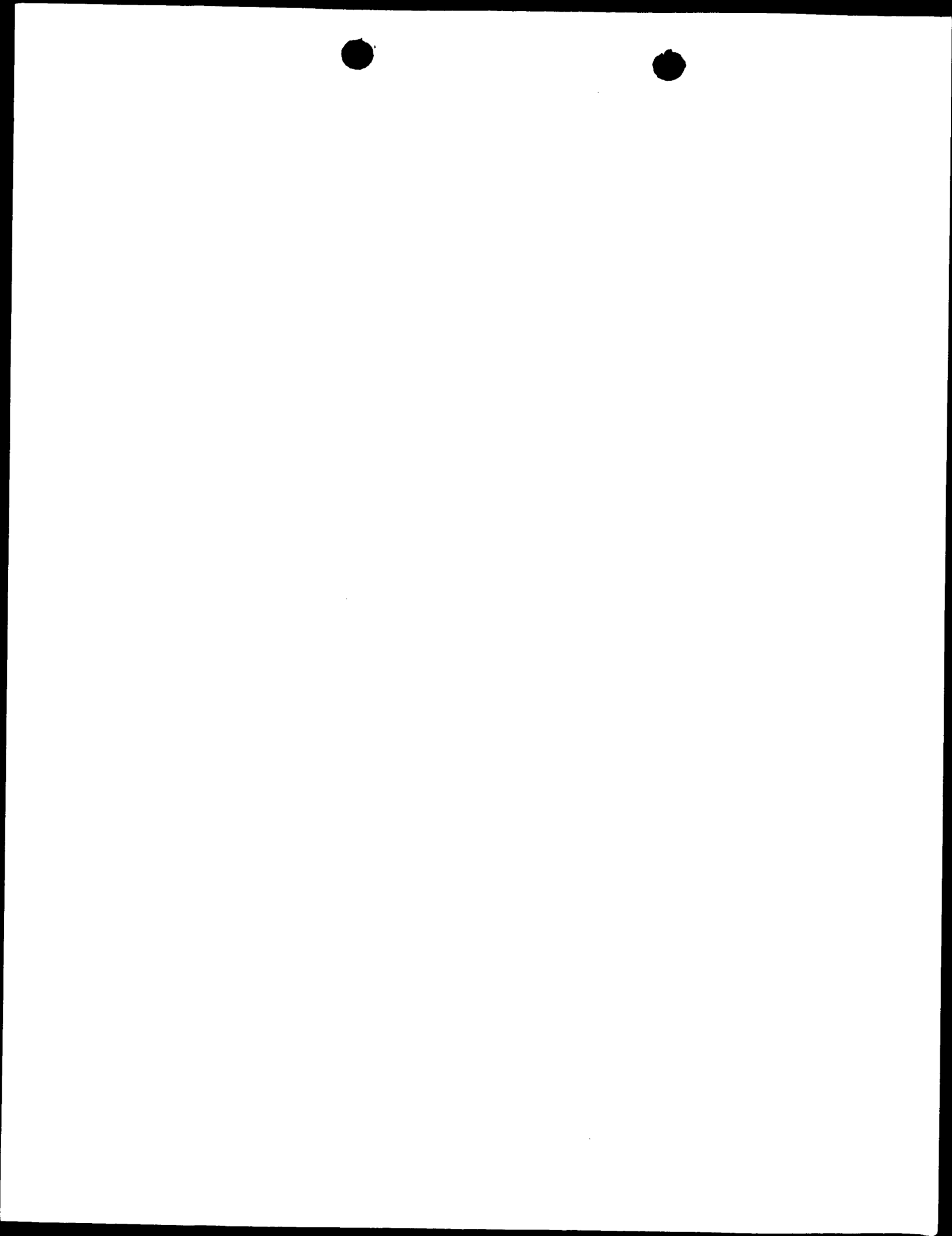
13-2-3	Validation messages Names	Green? Applicant 3.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Applicant 4.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Applicant 5.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Agent 1.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Agent 2.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Agent 3.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Agent 4.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.

PCT (ANNEX - FEE CALCULATION SHEET)

55791WO007

Original (for SUBMISSION) - printed on 03.08.2001 09:20:43 AM

		Green? Agent 5.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Agent 6.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
		Green? Agent 7.: Where several first/given names are indicated, they should preferably be separated by a comma. Please verify.
13-2-7	Validation messages Contents	Yellow! The power of attorney or a copy of the general power of attorney will need to be furnished unless all applicants sign the request form.
13-2-8	Validation messages Fees	Green? Please confirm that fee schedule utilized is the latest available
13-2-9	Validation messages Payment	Green? Please ensure that you have a valid deposit account with the receiving Office selected.



TRANSMITTAL LETTER TO THE UNITED STATES RECEIVING OFFICE

518 Rec'd PCT/PTO 0 8 AUG 2001

Date	August 8, 2001
International Application No.	PCT/US01/24867
Attorney Docket No.	55791WO007

I. Certification under 37 CFR 1.10 (if applicable)

EL596979466US
Express Mail Mailing Number

August 8, 2001
Date of Deposit

I hereby certify that the application/correspondence attached hereto is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Commissioner for Patents Washington, D.C. 20231.

<i>Carol Decaire</i>
Signature of Person Mailing Correspondence

Carol Decaire
Typed or Printed Name of Person Mailing Correspondence

II. ☒ New International Application

TITLE	PROCESS FOR PREPARING A MULTI-LAYER ARTICLE HAVING A FLUOROPLASTIC LAYER AND AN ELASTOMER LAYER	Earliest Priority Date (Day/Month/Year)
		23 AUGUST 2000

SCREENING DISCLOSURE INFORMATION: In order to assist in screening the accompanying international application for purposes of determining whether a license for foreign transmittal should and could be granted and for other purposes, the following information is supplied. (Note: check as many boxes as apply):

- A. ☐ The invention disclosed was not made in the United States.
- B. ☐ There is no prior U.S. application relating to this invention.
- C. ☒ The following prior U.S. application(s) contain subject matter which is related to the invention disclosed in the attached international application. (NOTE: priority to these applications may or may not be claimed on form PCT/RO/101 (Request) and this listing does not constitute a claim for priority.)

Application No.	09/644,731	filed on	23 AUGUST 2000
Application No.		filed on	

- D. ☒ The present international application ☒ is substantially identical to ☐ contains less subject matter than that found in the prior U.S. application(s) identified in paragraph C above.
- E. ☐ The present international application ☐ contains additional subject matter not found in the prior U.S. application(s) identified in paragraph C above. The additional subject matter is found on pages and ☐ DOES NOT ALTER ☐ MIGHT BE CONSIDERED TO ALTER the general nature of the invention in a manner which would require the U.S. application to have been made available for inspection by the appropriate defense agencies under 35 U.S.C. 181 and 37 CFR 5.1. See 37 CFR 5.15.

III. ☐ A Response to an Invitation from the RO/US. The following document(s) is (are) enclosed:

- A. ☐ A Request for An Extension of Time to File a Response
- B. ☐ A Power of Attorney (General or Regular)
- C. ☐ Replacement Pages:

pages		of the request (PCT/RO/101)	pages		of the figures
pages		of the description	pages		of the abstract
pages		of the claims			

- D. ☐ Submission of Priority Documents

Priority Document		Priority Document	
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- E. ☐ Fees as specified on attached Fee Calculation sheet form PCT/RO/101 annex

IV. ☐ A Request for Rectification under PCT Rule 91 ☐ A Petition ☐ A Sequence Listing Diskette

V. ☐ Other (please identify):

The person
signing this
form is the:

<input type="checkbox"/> Applicant	JAMES V. LILLY
<input checked="" type="checkbox"/> Attorney/Agent (Reg. No.) 27,817	Typed Name of Signer
<input type="checkbox"/> Common Representative	<i>James V. Lilly</i> Signature

PATENT COOPERATION TREATY

PCT

REC'D 04 DEC 2002

WIPO

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference F 3012 PCT	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/US01/24867	International filing date (day/month/year) 08/08/2001	Priority date (day/month/year) 23/08/2000
International Patent Classification (IPC) or national classification and IPC B29C47/06		
Applicant 3M INNOVATIVE PROPERTIES COMPANY et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 7 sheets, including this cover sheet.

- ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 1 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand 04/12/2001	Date of completion of this report 02.12.2002
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Lindner, T Telephone No. +49 89 2399 8976 

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/US01/24867

I. Basis of the report

1. With regard to the elements of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

Description, pages:

1-12 as originally filed

Claims, No.:

1-29 as originally filed

30 as received on 31/10/2002 with letter of 31/10/2002

Drawings, sheets:

1/1 as originally filed

2. With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/US01/24867

- ☐ the description, pages:
☐ the claims, Nos.:
☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims 1-30
	No:	Claims
Inventive step (IS)	Yes:	Claims
	No:	Claims 1-30
Industrial applicability (IA)	Yes:	Claims 1-30
	No:	Claims

- 2. Citations and explanations
see separate sheet**

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/US01/24867

Re Item I

Basis of the report

1. Reference is made to the following documents:

D1: US-A-5641445

D2: US-A-4895744

D3: WP-A-96/00657

2. The international application concerns processes for preparing multi-layer articles, more concretely, tubes or hoses comprising an elastomer layer extrusion coated with a fluoroplastic composition (claims 1, 28 and 29) and the product as such (claim 30).

The essence of the invention resides in thermally insulating the curable elastomer layer prior to contacting it with the molten fluoroplastic composition in order to prevent the adhesion force of being impaired in a subsequent curing step.

Re Item V

Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

- 3.1 D1 deals with the object of providing a multi-layered fuel tube made by coextrusion. Preferably, an inner layer of rubbery material such as FKM, which is a terpolymer of tetrafluoroethylene (TFE), hexafluoropropylene (HFP) and vinylidene fluoride (VF₂) and at least one outer layer of a plastic material such as THV, which also is a terpolymer of TFE, HFP and VF₂ (col.1, ll.20-29 of D1 and the disclosure of D3).

Such an article is known from D3, to which D1 explicitly refers.

Although D1 designates FKM a rubbery material, it emanates from D3 that FKM is a fluoroelastomer (p.5, ll.1-26). Otherwise, extrusion of FKM as envisaged by D1 would be difficult if not impossible.

THV is a fluoroplastic polymer - see D3 at page 6, line 5 et seq..

The materials envisaged in D1 and those contemplated by the present application at least partially overlap.

According to present claim 1, a precursor article comprises a curable elastomer layer to which a fluoroplastic layer is applied.

According to present claim 10, the elastomer comprises a fluoroelastomer.

D3 points to the incompatibility of FKM and THV in terms of extrusion temperature. In lines 22 to 24 at page 8, it is stated that the temperature of FKM is controlled in the extruder so that it does not overheat due to the relatively elevated temperature of the THV.

D1 addresses this problem.

In the paragraph bridging columns 1 and 2, it can be read that one problem to be solved is the difficulty in co-extruding a rubbery material with a plastic material due to the different extrusion temperature requirements of each during the process. If the rubber layer is subjected to temperatures of 480° F, the rubber layer will be scorched.

- 3.2 The solution to this problem provided by D1 is an insulation between the first die and the second die in order to minimize heat transfer there between (col.2, ll.21-23).

The problem of degradation of a layer of low melt flow temperature by the heat transferred from a layer of high melt flow temperature is realized by D2, too. As an alternative solution, it is suggested that an evacuated space be provided.

Art. 33(2) and (3) PCT

- 3.3 The fact that the present independent claims require a "precursor article" to be shielded from heat transfer whereas the extrusion process according to D1 focuses on a co-extrusion imparts novelty over D1.

D1 does not disclose insulation of the precursor article against heat transfer, but insulation of the die from which a curable elastomer is extruded against heat transfer from the die from which the fluoropolymer is extruded at a higher temperature.

3.4 This difference is not decisive for the assessment of inventive step.

Present claim 8 is directed to a step of extruding a curable elastomer composition through a die in order to form the precursor article.

There is no limitation as to the distance in space and/or time from the extrusion of the fluoropolymer this step should occur.

It might directly precede the coating of the fluoroplastic layer on the extruded elastomer layer.

The problem that the melt temperature of the fluoroplastic polymer exceeds the melt temperature of the elastomer to be coated therewith is the same in D1 and the present application.

Thermally insulating the curable elastomer layer prior to application of the fluoroplastic layer is suggested in both D1 and the present application.

The present application is silent as to when this step of insulating should start. From this fact, it cannot be derived that there is an approach which is different from that of D1, if the precursor article is a curable elastomeric layer which is preferably contacted for the first time with molten fluoroplastic in a crosshead die (present claim 3) and the means for protecting the curable elastomer is a sleeve positioned in an upstream opening of this die which receives the precursor article (present claim 4).

Moreover, a skilled person easily recognises that the problem underlying the present application and that one underlying D1 originate from the same cause, namely overheating the curable elastomer layer.

3.5 From D3, further features emanate such as steps (d) and (e) of present claim 1 and curability of the FKM inner layer - see claim 19 and the paragraph bridging pages 8 and 9 of D3.

A further outer layer which is extruded from a crosshead die and co-cured is also known from claims 1 and 11 of D3.

Bonding a third layer to the surface of the fluoroplastic layer the temperature of the former layer being also controlled is disclosed in D1 - see column 6, line 52 to column 7, line 56, particularly lines 38 to 56 at column 7.

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/US01/24867

In the light of this teaching, none of the present claims including claims 25 and 26 (cf. the adhesion test at the bottom of page 12 of D3) involves an inventive step.

4. A tube comprising an inner curable or cured layer bonded to an outer fluoroplastic layer represents one embodiment of a multi-layer article that can be obtained by the process of present claims 1 to 29.

Such an article is also obtained by the process disclosed in D1 without however indicating the bond strength between the respective layers.

If it is aimed at providing a material having an adhesion between an elastomer layer and a fluoroplastic layer of a minimum of 15 N/cm, this problem is not solved by claiming an article which has this desired property.

The solution to this problem as it is offered in the present specification is related to certain measures in the laminating step.

The gist of the invention lies in the manner according to which the layers are thermally isolated against each other in the crosshead of a die.

Thus, no inventive step is involved in the subject-matter of product claim 30.

-
30. A multi-layered article prepared by a process according to anyone of the preceding claims, said article comprising an elastomer layer and a flouroplastic layer, wherein the adhesion between said flouroplastic layer and said elastomeric layer is at least 15 N/cm.